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By M. E. HUBBARD † (Fellow)

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To the oil industry there is a further point of nearly equal importance, namely, the change in product-mix. In the Eastern Hemisphere gasolines represented some 33 per cent of total products in 1938 and only 23 per cent in 1952. In the U.S.A., however, the change was much less marked and dropped only from about 48 to 41 per cent over the same period.

In a paper presented to the Economics and Operations Group of the IP in January 1954,[1] the author examined this and showed that though the trend of demand towards the use of proportionately more fuels for static purposes was similar in both areas, its effect on the product-mix in the U.S.A. had been nearly offset by the still faster growth in the use of natural gas.

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The base year used in the Robinson Report was 1955, and the actual statistics for 1958 were available when the calculations were begun. A gas pipeline, *e.g.* from the Sahara, is unlikely to be in operation by 1965, but might possibly be so by 1968. The author has, therefore, taken 1958 and 1968 as convenient years to be considered.

SUMMARY OF CONCLUSIONS

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L

portion; gasoline itself will fall marginally, but gasolines plus LPG and refinery gas or gas-making components should increase slightly.

The major change will be between middle distillates and residuals. The former may reach 34 per cent of total fuel products (*cf* between 29 and 32 per cent during the last ten years) and residuals will fall to 40 per cent (*cf* 40 to 47 per cent previously).

METHOD OF ASSESSMENT

The object of the Robinson Commission was to forecast the future demand on primary energy resources, total coal, total oil, hydro-power, etc., and to what extent these could be available from indigenous sources as compared with imports. For this study the Commission had to give some consideration to the development of secondary forms of energy, notably manufactured gas and electricity, since these have a direct bearing on the primary energy requirements. Also the Commission received certain forecasts for the rate of growth of energy requirements in the main end-use sectors (iron and steel, other industry, road and air transport, etc.), but did not try to build up a complete picture of the future use of each form of energy in each sector.

This complete analysis is necessary if a forecast of petroleum product-mix is to be made.

The basic statistics are best available from the OEEC Annual Statistical Bulletins.[3] From these it is possible to follow primary energy through the manufacture of secondary energy and build up a complete flowsheet to the final end-use in each sector.

To estimate the future product-mix it is possible to reverse this procedure, starting by estimating the likely requirements of each sector in the light of its probable development, then to consider how secondary energy is likely to be obtained and using what sources within the framework of the primary energy assumed to be available.

Any forecast must be based on a number of assumptions. The principal ones adopted for this study are:

(1) Primary energy demand will rise broadly in line with the " mean " of the Robinson Report.

(2) No sudden and unforeseen change in fuel technology will occur.

TABLE I

Primary Fuels—1958

The first column under each heading is in actual units and the second column is the hard coal conversion factor.

	Hard coal		Lignite and briquettes		Crude oil		Natural gas		Hydro-power and geothermal		Nuclear		Total
	Million tons	Coal equiv. 1·0	Million tons	Coal equiv. 0·29	Million tons	Coal equiv. 1·43	10^9 m³ at 4200 cal/m³ §	Coal equiv. 0·60	10^9 kWh	Coal equiv. 0·40		Coal equiv.	Coal equiv.
Production (less stock changes)	444·0	444·0	111·3	32·4	12·5	17·9	15·6	9·4	176·7	70·7		0·1	571·5
Net imports primary fuels	+35·4	+35·4	+1·3	+0·4	+111·3	+159·0							
,, products	—	—	+4·9 *	+3·4	+13·4	+19·2							
Global availability	479·4	479·4	117·5	36·2	137·2	196·1	15·6	9·4	176·7	70·7		0·1	791·9‡
Less auto consumption	15·6	15·6	7·4	1·8	11·8†	16·9	0·7	0·4					
,, distribution losses							0·6	0·4					
Net availability	463·8	463·8	110·1	34·4	125·4	179·2	14·3	8·6	176·7	70·7		0·1	756·8
To conversion processes													
Oil refining					125·4	179·2							
Patent fuel	16·6	16·6											
Coke ovens	130·4	130·4											
Gasworks	39·5	39·5											
Electricity	106·5	106·5	47·2	13·7			1·7	1·0	176·7	70·7		0·1	
Total to conversion	293·0	293·0	47·2	13·7	125·4	179·2	1·7	1·0	176·7	70·7		0·1	557·7
To end-use sectors	170·8	170·8	62·9	20·7	—	—	12·6	7·6	—	—		—	199·1

* Actual weight of imported briquettes, conversion factor 0·69.
† Refinery fuel and loss plus non-fuel products, less substitute fuels.
‡ Excluding " others " given as 20 million tons HCE.
§ Equivalent to 472 Btu/cu ft.

(3) By 1968 a gas pipeline will be in operation importing some 15 million tons of coal equivalent (average about 12×10^8 cu ft/day) and that imports of liquefied natural gas will have risen to 3 million tons of coal equivalent. This in addition to the major increase in indigenous gas production to 30 million tons coal equivalent implied in the Robinson Report.

(4) Government policy, in general, is likely to give some protection to coal. It is therefore assumed that regardless of economic factors the quantity of coal given in the Robinson Report will in fact be used. The effect of any fiscal protection or compulsion is likely to be more effective in ensuring the use of coal in power stations and industry than in steel works and the domestic sectors.

(5) Quantities of other forms of indigenous energy are based on the Report with the necessary interpolations; this applies particularly to lignite and hydro-electricity.

TABLE II

Secondary Energy—1958

The first column under each heading is in actual units and the second column is the hard coal conversion factor.

	Patent fuel		Cokes				Gases								Petroleum										Electricity	
			Coke oven coke		Gasworks coke		Coke oven gas		Gasworks gas		Natural and refinery gas		Blast furnace gas		LPG		Gasolines		Kerosines		Gas/diesel oil		Fuel oil			
	Million tons	Coal equiv. 1·0	Million tons	Coal equiv. 0·96	Million tons	Coal equiv. 0·96	10^9 m³ *	Coal equiv. 0·60	10^9 m³ *	Coal equiv. 0·60	10^9 m³ *	Coal equiv. 0·60	10^9 m³ *	Coal equiv. 0·60	Million tons	Coal equiv. 1·43	Million tons	Coal equiv. 1·43	Million tons	Coal equiv. 1·43	Million tons	Coal equiv. 1·43	Million tons	Coal equiv. 1·43	10^9 kWh	Coal equiv. of fuels used
Source																										
Coal	17·0	17·0	130·4	130·4	39·5	39·5																			199·0	106·4
Lignite																									29·7	13·7
Oil									0·1	0·1	1·4	0·9														
Natural gas											16·8	10·0			2·1	3·0	26·3	37·6	5·0	7·2	31·3	44·8	59·2	84·7	27·3	10·6
Hydro and geothermal																									174·8 ‖	70·0‡
Coke ovens							45·3	27·2					51·9	30·9											20·0	9·8
Gasworks									21·6	13·0																
Nuclear																									0·3	0·1
Imports exports	−0·1	−0·1	+0·8	+0·7																						
Total	16·9	16·9	131·2	131·1	39·5	39·5	45·3	27·2	21·7	13·1	18·2	10·9	51·9	30·9	2·1	3·0	26·3	37·6	5·0	7·2	31·3	44·8	59·2	84·7	451·0	210·9 §
To other secondary forms																										
Coke oven gas			28·3†	27·2																						
Gasworks gas					13·5†	13·0									0·1	0·1										
Blast furnace gas			32·2†	30·9																						
Electricity							0·3	0·2			1·8	1·1	14·2	8·5									7·4	10·6		
Total			60·5	58·1	13·5	13·0	0·3	0·2			1·8	1·1	14·2	8·5	0·1	0·1							7·4	10·6		
Stock changes and adjustments			−3·4	−8·3	+3·2	+1·3																	−1·1	−1·5		
Less auto consumption	0·2	0·2	12·7	12·2	12·7	12·0	17·6	10·6	0·9	0·5	0·7	0·4	4·4	2·6											44·7	20·9
Less distribution losses							1·2	0·7	1·0	0·6	0·7	0·4	5·7	3·4											45·7	21·3
To end-use sectors	16·7	16·7	54·6	52·5	16·5	15·8	26·2	15·7	19·8	12·0	15·0	9·0	27·6	16·4	2·0	2·9	26·3	37·6	5·0	7·2	31·3	44·8	50·7	72·6	360·6	168·7
	← Total gases 10^9 m³ HCE 88·6 53·1 →																									

* Volume calculated at 4200 kg cal/m³.
† Coke equivalent.
‡ Hydro-electricity at 0·4 kg/kWh.
§ Average actual conversion factor, including hydro-electricity, was 0·47 kg/kWh.
" " " excluding hydro-electricity, was 0·51 kg/kWh.
‖ All gas fuels together.

1958 ENERGY BALANCE

Table I shows the primary energy listed in the OEEC estimates giving production and imports/exports and indicating what quantities passed to processes for their conversion into secondary energy and what quantities went direct to an end-use sector. On this and all subsequent tables the first of each pair of columns indicates the quantity as shown in the OEEC statistics, and the second expresses it in terms of coal equivalent.

Table II deals with secondary energy, tracing through the primary energy to the eventual secondary energy, including the various forms of manufactured gas and electricity. Natural gas is brought into this table, since some of it, at least, is mixed with manufactured gas. For the sake of convenience, oil products are considered as secondary energy and refining as a manufacturing operation.

Table III shows all the energy, primary or secondary, used in the seven sectors adopted by OEEC. The figures given derive directly from the statistics (with minor adjustments to take account of stock changes), except in so far as the division of gas/diesel and fuel oils are concerned; the total of each is available, but the split is the author's estimate.

TABLE III

End Uses by Sectors—1958

The first column under each heading is in actual units and the second column is the hard coal conversion factor.

Form of energy	Factor	Iron and steel		Other industrial		Road and air transport		Rail transport		Inland and coastal waters		Domestic and general		Ocean bunkers		Total	
		Units	Coal equiv.	Units	Coal equiv.	Units	Coal equiv.	Units	Coal equiv.	Units	Coal equiv.	Units	Coal equiv.	Units	Coal equiv.	Units	Coal equiv.
Coal	million tons 1·0	8·4	8·4	53·7	53·7	—	—	26·4	26·4	1·7	1·7	79·6	79·6	1·0	1·0	170·8	170·8
Patent fuel	million tons 1·0	0·1	0·1	0·4	0·4	—	—	1·8	1·8	—	—	14·4	14·4	—	—	16·7	16·7
Lignite	million tons 0·29	2·5	0·7	28·1	8·2	—	—	1·2	0·3	—	—	39·7	11·5	—	—	71·5	20·7
Coke oven coke	million tons 0·96	27·6	26·5	10·9	10·5	—	—	0·3	0·3	—	—	15·8	15·2	—	—	54·6	52·5
Gasworks coke	million tons 0·96	0·4	0·4	2·2	2·1	—	—	0·1	0·1	—	—	13·8	13·2	—	—	16·5	15·8
Gas (natural, coke oven, and gasworks)	10^9 m³ 0·60	14·1	8·5	21·4	12·8	(0·7)	0·4	—	—	—	—	23·6	14·2	—	—	59·8	35·9
Blast furnace gas	10^9 m³ 0·60	27·1	16·1	0·5	0·3	—	—	—	—	—	—	—	—	—	—	27·6	16·4
LPG	million tons 1·43	—	—	0·3	0·4	—	—	—	—	—	—	1·7	2·4	—	—	2·0	2·8
Gasoline	million tons 1·43	—	—	0·2	0·3	24·8	34·5	—	—	0·1	0·1	1·2	1·7	—	—	26·3	36·6
Kerosine	million tons 1·43	—	—	0·1	0·1	1·5	2·2	—	—	—	—	3·4	4·9	—	—	5·0	7·2
Gas/diesel oils	million tons 1·43	—	—	2·0	2·9	9·6	13·7	2·3	3·3	3·0	4·3	11·0	15·7	3·4	4·9	31·3	44·8
Fuel oils	million tons 1·43	4·7	6·7	24·0	34·3	—	—	—	—	0·6	0·8	11·4	16·5	10·0	14·3	50·7	72·6
Electricity	10^9 kWh 0·47	33·3	15·7	189·8	89·3	—	—	16·9	7·9	—	—	123·2	58·0	—	—	363·2	170·9
			83·1		215·3		50·8		40·1		6·9		247·3		20·2		663·7

FORECASTING OF END USAGE

The following are some of the main considerations which led to the establishment of Table IV:

Iron and Steel

The increase in energy requirements during the ten years 1958–68 is taken as 47½ per cent (equivalent to 4 per cent pa). Owing to possible technical developments, one half of the increased output of blast furnaces is considered to be using natural gas by 1968. Electricity consumption is taken as doubled and fuel oil as fully tripled at the absolute cost of coal and gasworks coke and at the relative cost of blast furnace gas.

Other Industry

Increased total demand taken at 33 per cent (3¼ per cent pa). Use of electricity increased by 75 per cent (equivalent to 5¾ per cent pa). Gas, including natural gas, is likely to be much more freely available and, owing to its convenience, it would seem reasonable to suppose a major increase in consumption; this has been taken as 31 million tons coal equivalent as compared with 12·8 million tons in 1958, equivalent to an average rise of 9 per cent pa. Lignite consumption is likely to rise, but coke oven and gas coke are assumed to remain constant. By difference, the quantity of the remaining forms of energy, namely, coal and oil, is actually 5 million tons coal equivalent less than in 1958. Industry may, as mentioned above, be under some pressure to use coal, but the advantages of oil are such that the swing to that fuel will continue, though at a lower rate than in the past. It is therefore assumed that oil consumption will only increase from 26 million to 30 million tons. Coal will have to accept a material reduction from 53·7 million to around 41 million tons.

Road and Air Transport

Increased total demand 70 per cent (equivalent to 5½ per cent pa). The swing to the diesel on the road can be expected to continue, but less rapidly; the present ratio gasolines/diesel oil is 2·5 : 1, and it is assumed that this will drop to 2·25 : 1 by 1968. Avgas will become of secondary importance, but aviation turbine fuel may well be four times greater, predominantly of ATK.

Rail Transport

Total requirements will fall by 20 per cent owing to electrification and dieselization. Coal consumption is assumed to drop from 26·4 million tons to 10 million tons, but both electricity and diesel oil consumption to be roughly doubled.

Inland Waterways and Coastal Vessels

Only a small increase in requirements, say 20–25 per cent. Virtually all will be for diesel oil.

Domestic and Miscellaneous

There is no way of gauging what proportion of this very important sector is in fact domestic. Agriculture represents a certain, though small, part, and the heating of commercial and industrial premises is undoubtedly significant. However, OEEC does not appear to expect an overall increase in consumption much greater than the increase in population. For this study the increase taken is $1\frac{3}{4}$ per cent pa or 17 per cent over the ten year period. On the other hand, this is a sector where the type of energy used is likely to change greatly, gas and electricity limiting the gain in oil and drastically reducing the demand for solid fuels. Over the last ten years the use of electricity has been rising at an average of 9·5 per cent pa; this rate may fall somewhat to, say, 7 per cent in future. Gas demand has been rising at $3\frac{1}{4}$ per cent pa and with increased supplies and the effort to sell natural gas, a growth of 4 per cent pa in future seems reasonable. LPG, still a small contributor but rising fast, may retain its impetus and more than double.

Oil consumption has been rising very rapidly in the recent past, at over 20 per cent pa during the last ten years and still at 14 per cent during the last three years. This rapid rate must slow up eventually, particularly since solid fuel cannot be completely displaced and gas can eventually be expected, as in the U.S.A., to replace oil on account of its convenience. Over the ten year period a rise of two-thirds has been assumed for oil, $5\frac{1}{2}$ per cent pa on the average, though most of the growth would take place in the early part of the period.

These suppositions mean a reduction in solid fuels from 124 million tons coal equivalent to 96 million tons; broadly, all this would be at the cost of coal (raw) with only minor changes in lignite, briquette, and coke consumption.

Ocean Bunkers

A 3 per cent pa average increase in consumption is assumed, equal to 34 per cent over ten years. In 1958 20 per cent of bunkers was diesel oil and 80 per cent was fuel oil. The swing towards the heavier fuel is likely to continue, though more slowly than in the past, and 15 per cent of diesel oil seems a likely proportion in 1968.

TABLE IV

End Uses by Sectors—1968

The first column under each heading is in actual units and the second column is the hard coal conversion factor.

Form of energy		Factor	Iron and steel		Other industrial		Road and air transport		Rail transport		Inland and coastal waters		Domestic and general		Ocean bunkers		Total	
			Units	Coal equiv.	Units	Coal equiv.	Units	Coal equiv.	Units	Coal equiv.	Units	Coal equiv.	Units	Coal equiv.	Units	Coal equiv.	Units	Coal equiv.
Coal	million tons	1·0	5·0	5·0	41·0	41·0	—	—	10·0	10·0	0·5	0·5	46·0	46·0	—	—	102·5	102·5
Patent fuel	million tons	1·0	—	—	—	—	—	—	—	—	—	—	15·0	15·0	—	—	15·0	15·0
Lignite	million tons	0·29	1·7	0·5	53·5	15·5	—	—	—	—	—	—	45·0	13·0	—	—	100·0	29·0
Coke oven coke	million tons	0·96	34·4	33·0	11·0	10·5	—	—	—	—	—	—	8·3	8·0	—	—	53·7	51·5
Gasworks coke	million tons	0·96	—	—	2·1	2·0	—	—	—	—	—	—	14·6	14·0	—	—	16·7	16·0
Gas (natural, coke oven, and gasworks)	10^9 m³	0·60	20·8	12·5	51·7	31·0	—	—	—	—	—	—	35·0	21·0	—	—	107·5	64·5
Blast furnace gas	10^9 m³	0·60	34·2	20·5	—	—	—	—	—	—	—	—	—	—	—	—	34·2	20·5
LPG	million tons	1·43	—	—	1·4	2·0	0·7	1·0	—	—	—	—	5·6	8·0	—	—	7·7	11·0
Gasoline	million tons	1·43	—	—	0·4	0·5	37·0	53·0	—	—	—	—	1·4	2·0	—	—	38·8	55·5
Kerosine	million tons	1·43	—	—	—	—	5·6	8·0	—	—	—	—	3·5	5·0	—	—	9·1	13·0
Gas/diesel oils	million tons	1·43	—	—	2·1	3·0	16·8	24·0	4·9	7·0	5·2	7·4	25·8	37·0	2·8	4·0	57·6	82·4
Fuel oils	million tons	1·43	15·4	22·0	28·0	40·0	—	—	—	—	0·4	0·6	11·9	17·0	16·2	23·0	71·9	102·6
Electricity	10^9 kWh	• 0·425	67·0	29·0	333·0	141·0	—	—	32·0	14·0	—	—	244·0	103·0	—	—	676·0	287·0
				122·5		286·5		86·0		31·0		8·5		289·0		27·0		850·5

• Estimated all-in average factor, including hydro-electricity, assuming 4 per cent gain in average thermal generating efficiency in ten years.

Electricity Generation

Total electricity production, in kWh, is likely to rise by 85 per cent, equivalent to, say, $6\frac{1}{2}$ per cent pa. Whatever hydro-electricity is produced will be used, but with the increasing cost of developing new schemes hydro output will not rise as fast as total electricity demand. Electricity generated by thermal stations will therefore rise by 110 per cent ($7\frac{3}{4}$ per cent pa). However, the increase in fuel consumption will not be as much owing to the improvement in efficiency.

The conversion of electricity into terms of coal equivalent is always a vexed question. The Robinson Commission adopted throughout their report a coefficient of 0·40 kg of coal per kWh. This, equivalent

TABLE V

Secondary Energy—1968

The first column under each heading is in actual units and the second column is the hard coal conversion factor.

	Patent fuel		Cokes				Gases						Petroleum										Electricity ‡	
			Coke oven coke		Gasworks coke		Coke oven gas		Gasworks gas		Natural and refinery gas		Blast furnace gas		LPG		Gasolines		Kerosines		Gas/diesel oil		Fuel oil			
	Million tons	Coal equiv. 1·0	Million tons	Coal equiv. 0·96	Million tons	Coal equiv. 0·96	10^9 m³ *	Coal equiv. 0·60	10^9 m³ *	Coal equiv. 0·60	10^9 m³ *	Coal equiv. 0·60	10^9 m³ *	Coal equiv. 0·60	Million tons	Coal equiv. 1·43	Million tons	Coal equiv. 1·43	Million tons	Coal equiv. 1·43	Million tons	Coal equiv. 1·43	Million tons	Coal equiv. 1·43	10^9 kWh	Coal equiv. of fuels used
Source																								
Coal	15·2	15·2	124·5	124·5	41·9	41·9																		379·0
Lignite																								48·0
Oil											3·4	2·0			7·7	11·0	40·2	57·5	9·1	13·0	57·6	82·4	78·9	112·6	23·0	10·0
Natural gas											75·0	45·0													22·0	9·7
Hydro and geothermal																									262·0	105·0
Coke ovens							49·1	29·5																		
Gasworks									22·8	13·7															24·0	10·5
Blast furnace gas													64·2	38·5												
Nuclear																									75·0	30·0
Total	15·2	15·2	124·5	124·5	41·9	41·9	49·1	29·5	22·8	13·7	78·4	47·0	64·2	38·5	7·7	11·0	40·2	57·5	9·1	13·0	57·6	82·4	78·9	112·6	833·0	353·7
To other secondary forms																										
Coke oven gas			30·7†	29·5																						
Gasworks gas					14·3†	13·7											1·4	2·0								
Blast furnace gas			40·0†	38·5																						
Electricity											16·2	9·7	17·5	10·5									7·0	10·0		
Total			70·7	68·0	14·3	13·7					16·2	9·7	17·5	10·5			1·4	2·0					7·0	10·0		
Stock changes and adjustments																										
Less auto consumption	0·2	0·2	5·2	5·0	12·7	12·2	19·3	11·6	0·8	0·5	—	—	7·5	4·5											72·0	30·5
Less distribution losses							1·3	0·8	1·0	0·6	4·2	2·5	5·0	3·0											85·0	36·2
To end-use sectors	15·0	15·0	53·7	51·5	16·7	16·0	28·5	17·1	21·0	12·6	58·0	34·8	34·2	20·5	7·7	11·0	38·8	55·5	9·1	13·0	57·6	82·4	71·9	102·6	676·0	287·0
							← Total gases 10^9 m³ HCE 141·7 85·0 →																			

* Volume calculated at 4200 kg cal/m³.

† In coke equivalent.

‡ Hydro and nuclear power at factor 0·40. Thermal power at factor 0·435.

to 30 per cent efficiency in a thermal generating station, was taken as a figure likely to be reached by 1975. Analysing the 1958 electricity figures, and leaving hydro-electricity at 0·40 coefficient, the actual consumption of all thermal stations was 0·51 kg per kWh (23½ per cent efficiency). The overall efficiency of generating in the U.K. improved by 5 per cent over the last ten years,[4] in part due to the replacement of much over-age equipment which was still in service at the beginning of the period. Such a rate of progress does not seem possible in future,[5] and a 4 per cent improvement is more likely over the next ten years. For the purposes of this study, therefore, whilst the efficiency of hydro-electricity is assumed to remain constant at 0·40 kg, that of thermal generation in 1968 is taken as 0·435 kg (27½ per cent efficiency).

1968 ENERGY FLOW SHEET

Based on the above considerations, and accepting broadly the Commission's forecast of availability, an attempt has been made to establish a set of tables similar to those worked out for 1958.

Table IV starts with the end-use sectors.

Table V represents a possible means of providing the secondary energy needed by these sectors.

Table VI brings it back to primary fuels.

The forms of energy used for electricity generation have been discussed above.

As regards gas, the activity of coke ovens is set mainly by the steel industry. It has been assumed that gasworks will, on the whole, contribute about the same as in 1958; production is likely to increase in areas near coal-fields and remote from a gas pipeline

TABLE VI

Primary Fuels—1968

The first column under each heading is in actual units and the second column is the hard coal conversion factor.

	Hard coal		Lignite and briquettes		Crude oil		Natural gas		Hydro-power and geothermal		Nuclear		Total
	Million tons	Coal equiv. 1·0	Million tons	Coal equiv. 0·29	Million tons	Coal equiv. 1·43	10^9 m³ *	Coal equiv. 0·60	10^9 kWh	Coal equiv. 0·40	10^9 kWh	Coal equiv.	Coal equiv.
Global requirements	466·6	466·6	181·0	52·5	226·9	324·5	80·0	48·0	262·0	105·0		30·0	1,027
Less auto consumption	15·0	15·0	8·5	2·5	15·0	21·5	0·8	0·5					
,, distribution losses							4·2	2·5					
Net requirements	451·6	451·6	172·5	50·0	211·9	303·0	75·0	45·0	262·0	105·0		30·0	985
To conversion processes													
Oil refining					211·9†	303·0							
Patent fuel	15·2	15·2											
Coke ovens	124·5	124·5											
Gasworks	41·9	41·9											
Electricity	167·5	167·5	72·5	21·0			75·0	45·0	262·0	105·0	75·0	30·0	
Total to conversion	349·1	349·1	72·5	21·0	211·9	303·0	75·0	45·0	262·0	105·0		30·0	853
To end-use sectors	102·5	102·5	100·0	29·0	—	—	—	—	—	—		—	131

* Calculated at 4200 kg cal/m³.

† Total oil fuel products 193·9 plus 15 million tons non-fuel products.

In hard coal equivalent the increase in fuel for thermal and nuclear power stations is therefore likely to increase from 141 million to 249 million tons, equivalent to 6 per cent pa. Owing to the cheapness of lignite in certain areas, all the production not used for other purposes will be burnt in power stations. Secondly, it can be assumed that the result of Government coal policy will mean that all coal not used for other purposes will go to electricity generation. Surplus natural gas will go there also under interruptable supply contracts. Any nuclear electricity will have to be absorbed, but this will still be a minor factor in 1968.

Oil will only be used in so far as the above are insufficient, and it would appear that the consumption in 1968 will be no greater, possibly even less, than in 1958.

or seaport but decrease in other areas. Further, an increase in transfers of refinery gas (or gasoline fractions) to gasworks is probable. The main increase in gas supply will, of course, be from natural gas, and of this possibly two-thirds from indigenous sources. Whether this natural gas is handled separately or in mixture with manufactured gas is immaterial for the purposes of this study.

In working out the tables the main problem was to bring the demand requirements in line with primary energy availability. The main place where there is (at least in the medium/long term) a wide margin of flexibility in form of energy used is in electricity generation. The balance had to be met there, and the calculations, as well as common sense, showed that much increased quantities of coal would have to be used.

CONCLUSIONS

The demand for products in 1958 and the forecast for 1968 is as follows:

	Million tons oil		Increase, average % pa
	1958	1968	
Refinery gas *	1·1	3·4	12
LPG	2·1	7·7	14
Gasolines	26·3	40·2	4·4
Kerosines	5·0	9·1	6·1
Gas/diesel oil	31·3	57·6	6·3
Fuel oil	59·2	78·9	2·9
Others	3·6	15·0	12
Total products	128·6	211·9	
Refinery fuel and loss	8·6	15·0	
Total crude oil	137·2	226·9	5·2

* Or light gas-making fractions.

The question of non-fuel products is a difficult one to resolve. The figure for 1958 given in the OEEC statistics seems to be low, but had, for arithmetical reasons, to be accepted for this study. With the rapid increase in petroleum chemicals, etc., a 1968 requirement of 15 million tons has been assumed, but no attempt made to forecast from which part of the barrel it would originate.

Expressed as percentages of total fuel products demand, the above figures are as follows:

	% fuel products	
	1958	1968
Refinery gas	0·9	1·7
LPG	1·7	3·9
Gasoline	21·0	20·4
Kerosine	4·0	4·6
Gas/diesel oil	25·0	29·3
Fuel oil	47·4	40·1
	100·0	100·0

These may usefully be put in perspective by comparing them with the recent figures for Western Europe.

TABLE VII

	% on total fuel products consumption					
	1950	1952	1954	1956	1958	1968
Refinery gas	—	—	0·3	0·3	0·9	1·7
LPG	0·6	1·0	1·3	1·5	1·7	3·9
Gasolines	28·2	26·6	25·4	23·0	21·0	20·4
Total gasolines and lighter	28·8	27·6	27·0	24·8	23·6	26·0
Kerosines	6·2	5·7	4·9	4·5	4·0	4·6
Gas/diesel oil	24·0	26·7	25·9	26·4	25·0	29·3
Total middle distillates	30·2	32·4	30·8	30·9	29·0	33·9
Fuel oil residuals	41·0	40·0	42·2	44·3	47·4	40·1
	100·0	100·0	100·0	100·0	100·0	100·0

From this it would appear:

1. That though the gasoline proportion itself will continue to fall marginally, this will be more than compensated by the sales of refinery gas (or gasoline components) to gasworks and by LPG. The total " gasolines and lighter " group should show a small increase in percentage.

2. Fuel oil or residuals will drop considerably to as low a percentage as we have known in the last ten years.

3. Middle distillates will rise to a proportion considerably higher than in the past, kerosine remaining roughly constant and the whole of the increase being concentrated in gas/diesel oil, particularly of the heating grades.

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DISCUSSION

C. P. Dalton (Esso Petroleum Co. Ltd): I have two broad comments on the basis of the figures themselves, the first of which concerns the year 1968. Not that I have any quarrel with the choice of this particular year—it seems as good as any other in a survey of this sort. It is, however, a point in time—a snapshot rather than a moving picture. The mental projection I have of 1968 is of proportionately large increases in natural gas and hydro and nuclear power at the expense of the growth rate for fuel oil. But I think it important to establish a clearer picture of the trend of fuel oil development in the years before and after 1968.

My second point concerns the assumptions on which the 1968 demand for fuel oil is based. These are made explicit in Mr Hubbard's calculations, and it is interesting to draw them together:

(1) A fixed Robinsonian framework of primary energy demand.

(2) A protected market and stable production for coal.

(3) Surplus coal and natural gas, and nuclear power, to be absorbed in the electricity industry at the expense of oil.

Fuel oil is, in fact, the balancing item irrespective of considerations of choice or price, and the electricity

industry is the main fulcrum at which pressure is applied. This may well be practical politics from an overall viewpoint. However, history is made by individuals; the extent to which the individual firm in the oil industry, especially the aggressive marketer trying to increase his volume, will review its actions in the light of Mr Hubbard's assumptions is a very open question. Sometimes the lumberjack cannot see the wood for the trees!

The important point that stems from these assumptions is, I think, that Mr Hubbard's longer-term estimates for fuel oil are subject to very considerable variation, *e.g.* if men get tired of earning a living underground and the assumed long-term coal production figures are not achieved. With this reservation, I accept the author's broad conclusion that the growth rate for fuel oil may tail off and that for middle distillates will accelerate. I should now like to touch on one of the main implications of this prediction.

If residual oil is to be the balancing fuel, its demand will surely become more subject to short-term fluctuations caused by the vagaries of the weather and the business cycle. To the extent that dual firing becomes more prevalent in Europe, these fluctuations will become even more pronounced. The oil industry at large has a reputation for flexibility in meeting such problems, but those in the industry who are particularly concerned with operations will appreciate what this means in practical terms, such as capacity in tankers, trucks, storage, and supplies and the costs of having these available.

To some extent, operating problems will be eased by the fact that the pattern of product demand suggested for 1968 seems more consistent with the yield from the lighter Saharan and Libyan crudes which will be coming to Europe in rapidly increasing quantities in the years ahead. The continuation of Europe's present position as a sizeable marginal importer of fuel oil would help to spread the load.

M. E. Hubbard: I agree with practically all Mr Dalton's points, particularly that of wanting a clearer trend of fuel oil development before and after 1968. $\frac{\delta Q}{\delta T}$ is what we would like, but I am afraid I have forgotten all my calculus.

Mr Dalton referred to coal miners getting tired of working underground. We think they ought to get tired, but they do not seem to. Practically the only place that I know where coal miners are willing to give up that life is in the Ruhr, where they can get industrial jobs right alongside. In France several coalfields are scheduled to be closed by the Coal and Steel Community, but the miners will just not accept it. The French Government has been forced to keep those mines going, because they fear bloodshed if they try to shut them down.

S. K. Gold (Caltex Services Ltd): In other talks I have heard, Mr Hubbard has made quite a point of the tax angle on petrol being a hidden subsidy for diesel fuel and the diesel engine. I wonder what effect this may have in the coming decade.

M. E. Hubbard: I did mention the assumption in the paper, namely, that the swing to diesel on the roads can be expected to continue, though less rapidly. The present ratio of gasolines to diesel oil is 2·5 : 1, and I assumed that by 1968 this will drop to about 2·25 : 1. There are some signs that the move to the diesel engine in the smaller vehicle is slowing up. There is not such a rapid swing towards the diesel as there was in the past, though the change of ratio I have assumed does represent quite a material cumulative change. I consulted some diesel experts on it, and they thought it was as good a guess as anybody dared make at the present moment. It has, of course, a material bearing on the overall product-mix.

J. G. Withers (BP Trading Ltd): I am very surprised that the domestic consumption of kerosine is anticipated to remain constant. If it is assumed that portable stoves will use the same amount of fuel that they do at the moment, we have in addition a very large field in flued convectors, domestic water-heating boilers, and the smaller-sized central heating stoves, all using kerosine, and I hope that there will be a considerable expansion there.

M. E. Hubbard: Yes, in the U.K. But remember that we are the most backward country in Western Europe in domestic heating. Other countries have passed through the heater stage and are now centrally heated on a much bigger scale. Hence kerosine is being driven out in many European countries by gas oil. In the U.K., also, there has been a very big improvement in burners in the last few years. I know of several cases right now where people are changing from kerosine pot burners to pressure burners using gas oil. As I see it, the gain in kerosine used for space heating is being largely if not entirely offset by the subsequent switch to gas oil for full central heating.

W. L. Newton (Petroleum Economics Ltd): There are two points which I should like Mr Hubbard to develop further. The first relates to the industries in which the use of fuel oil may actually decline at a later stage after reaching a peak. Mr Hubbard has suggested electricity. Are there any other industries which he would suggest where oil might be replaced by coal or natural gas at a later stage?

My second point relates to the increase in LPG consumption. Mr Hubbard has suggested that LPG consumption will go up by 14 per cent per annum. We all know that the LPG consumption by certain countries on the Continent, such as Italy and France, is on a very much greater scale than, for instance, in the U.K. Does Mr Hubbard believe that the use of LPG will also spread in countries such as the U.K? If so, does he believe this will be for reasons of ease of handling, or for other, *e.g.* economic, reasons, and in which end-use sector? I know that his figures show that the domestic use will go up fastest, but does he also think of petroleum chemicals, and in which other uses does he think LPG will be employed?

M. E. Hubbard: Industries with a reduction in fuel oil? Individual industries, yes, but I was not able to consider individual industries, as I started from the Robinson Report and the OEEC statistics, which do not distinguish between the iron and steel industries and all other manufacturing industries together. There are certainly some where there will be a reduction in the use of oil. I can think of glass manufacture and heat treatment in the metal industries, which are certainly moving away towards gas and electricity. But I do not think, if one takes other industries as a whole, one will obtain a reduction in fuel oil. It may not increase so rapidly, because a lot of the future increase will be taken by gas and electricity.

Concerning LPG, I do not know the answer. I was guided to some extent by my own experience. In the early thirties I built the first LPG manufacturing plant in

France, maybe in Europe, and we all thought that by the following year we would have reached the ceiling of LPG demand. How wrong! It has gone on growing ever since at a practically ever-increasing rate. It is natural to say that "it can't go on growing like this for ever," but so far no country has ever shown signs of reaching saturation level. The U.K. has a fantastically low consumption of LPG; I know there are better electricity and gas distribution systems here than in other countries, but I cannot help believing that some day people are going to find the convenience factor of LPG greater than they thought. Just now it is increasing very rapidly in Scandinavia. I am afraid I took a more or less cockshy guess based on previous rates of expansion.

B. A. Rahmer (Petroleum Press Service): I would like to comment on two assumptions in this paper. One is the rise in the natural gas availability which will no doubt take place; but I would like to draw your attention to the quite recently published report of the Gas Committee of the OEEC where the forecast previously made by the Energy Committee, practically identical with the Robinson people, has been somewhat scaled down, and the paper also said quite explicitly that in spite of the probability of a great increase in the availability of natural gas, this will almost certainly not make any strong influence on the energy supply—the whole make-up of energy in Europe within the 1960's.

The other thing is the question of coal. The question of the coal miners in the Ruhr has already been mentioned, but it seems to me that this is not a single case. It is the biggest concentration of coal mining on the Continent, and the situation in this respect has changed significantly since the Robinson Report. At that time, especially in Germany, perhaps the most urgent economic problem was the coal crisis, and now it is largely the labour crisis. It is accepted in Germany, and to some extent in Belgium, that coal production has to be somewhat reduced. In the U.K. the question of coal production has always been, to some extent, a matter of party politics. We can see that the present Government has re-affirmed its statement against protection, and there is at the moment not a very strong likelihood of this Government being overthrown, so altogether it seems to me that the trend since the Robinson Report has been published, and even more so since the time that the Robinson people discussed the matter, has been rather away from protectionism.

The final point is that, as already mentioned, fuel oil has, after all, gone up very considerably in the past two years, and the fuel oil consumption can now be not very far below the figure given in Mr Hubbard's paper for 1968. The implication would therefore be that if we accept the 1968 proportions, for the next few years there will be practically no increase in fuel oil; and yet we find even in a country such as Italy, where gasoline consumption is rising very rapidly, that the government has just now issued a forecast, according to which fuel oil will continue to increase, in the 1960's, at a higher rate than any other petroleum product.

M. E. Hubbard: Mr Rahmer points out that the OEEC Gas Committee has rather toned down the part of the Robinson Report on natural gas availability. I agree that the Robinson forecast does look pretty big. But if I did not accept the Robinson figures I could not have made the calculations. My calculations were based on the Robinson Report and if, in our particular case, I had substituted some other figure, I would have falsified the whole of the basis of the calculation.

With regard to coal, Mr Rahmer suggests that several of the main coal-producing countries now seem less protective minded than when the Robinson Committee made its study. I would remind you that in Germany a tax of 25 DM was imposed after the Robinson Committee worked on the subject. I admit it was some time later that the report was published, but the base work was done nearly a year before the report itself saw the light of day.

The position with regard to fuel oil has changed a lot since the base year of 1958, but again that had to be my starting point. It comes to this: if fuel oil goes on rocketing, will there not be arbitrary action by governments to restrict it? I do not see how we can go on with a rocketing fuel oil economy, with the coal position as it is in Europe, and be left in peace.

H. E. Barry (Shell-Mex and B.P. Ltd): I was a little surprised at Mr Hubbard's reply to Mr Withers' question about kerosine, but I think that here we might be up against a question of definition. Has stove oil or range oil of the quality used in Europe been included under gas/diesel oil or kerosine? If the Robinson Report treats the product used in the vaporizer type domestic boiler as kerosine, which in fact it is, I can see the market for this increasing quite appreciably. Another factor which I should have thought would have increased kerosine consumption considerably is the rapid progress in civil aviation and the growing use of Avtur—again a product in the kerosine range. After the recent efforts of Lord Brabazon, I am rather doubtful whether we shall see any large switch over to wide-cut gasoline, at least by British operators. With these two developments I have mentioned, I should like to ask Mr Hubbard whether he is happy with the modest increase shown for kerosine?

With regard to Mr Newton's question on LPG, perhaps I might supplement Mr Hubbard's reply. I feel that there is a large future for LPG industrially; although the market is relatively small, there has been a very large increase in the U.K. over the last two or three years. It has been mainly in the industrial field, with very little in the domestic field. In addition, there is also quite a heavy demand by the Gas Boards for butane for enriching coal gas, and with the difficulty of getting good gas-making coal this is an outlet which seems likely to grow. Hence I agree with Mr Hubbard that the future for LPG looks very promising.

M. E. Hubbard: I did not worry too much about the distinction between kerosine and gas oil, because I was aiming at estimating middle distillates. After all, from a refining angle there is such flexibility between kerosine and gas oil that it is rather gilding the lily to try to distinguish carefully between those two products. From the point of view of the broad trends in the industry, the three products which count are gasolines, middle distillates, and residuals. As regards aviation turbine fuel, I did assume that it would increase four times in the ten years.

W. Jamieson (British Petroleum Co. Ltd): I am in rather an embarrassing position in that for many years I worked under Mr Hubbard and now find myself rising to criticize some of the detail in his paper. It has been made clear, however, that Mr Hubbard has prepared this paper entirely in his private capacity.

Some of us have been rather puzzled by certain of Mr Hubbard's assumptions, as well as his arithmetic. One particularly puzzling assumption is that he foresees little increase in the consumption of fuel oil in the domestic and general sector. I believe this is a question of

definition and that in fact the original data upon which Mr Hubbard's analysis is based includes under fuel oil the extra light fuel oil which is sold in some countries and which from a product-mix point of view is essentially a middle distillate. Making an adjustment for this in Mr Hubbard's forecast would reduce his middle distillate requirement and increase his fuel oil requirement in 1968 by some two percentage points, and taking the alternative forecast which he has just given us, this adjustment would make the 1968 product-mix only marginally different from the current position.

There are other items which one could challenge in the author's detailed analysis, but I think the essential feature of the paper is that the analysis is only one of many possible situations which could be derived from Robinson's Report. I was rather surprised to hear Mr Hubbard say that his estimate is based generally upon the mean of Robinson's supply figures. A strict interpolation of Robinson's total energy forecast would give a mean figure for demand in 1968 of 1096 million tons and a low estimate of 1045 million tons. Mr Hubbard's estimate is 1047 million tons, *i.e.* virtually on Robinson's low. On the other hand, his estimate of 30 million tons coal equivalent of nuclear energy and 48 million tons of natural gas are both at the high of Robinson's estimate. The effect of this selection of estimates is, as Mr Dalton has already pointed out, to reduce the possible demand for fuel oil, and, indeed, Mr Hubbard's estimated fuel oil demand in 1968 would appear to be no higher than demand to-day, and his total oil demand is marginally lower than the low forecast given by Robinson.

Mr Hubbard may well be right, although I think it is unrealistic to assume such a high increase in nuclear energy in the circumstances which he is forecasting of apparent surplus energy. With regard to natural gas, while there may well be a pipeline from the Sahara in 1968, it does not necessarily follow that it will be running at full capacity.

The real point of all this is that everybody in this room could take the range of supply estimates given by Robinson and each of them could work out entirely different answers in terms of product-mix. This is another way of saying that the oil industry cannot be certain of the fuel oil demand in the next decade, nor assured of any special trend in the product-mix.

The lesson from this is that the industry is going to need not less but probably more flexibility in the future. I think we do not make enough of the importance of flexibility to the oil industry and that it is worth saying, looking back on the last decade, that if the oil industry had not been flexible, the European economy would not be as buoyant as it is to-day. Looking to the future, if there is going to be a restriction on the activity of the oil industry through some form of protectionism for indigenous fuels, as implied in Mr Hubbard's analysis, then there will be a danger of Europe's economic progress being hindered by this loss of flexibility.

M. E. Hubbard: I certainly deserve all the remarks Mr Jamieson made and could not agree more that every one of us, given the same data, would reach different conclusions. Mr Jamieson started off by comparing my total energy in 1968 with the mean of Robinson. I hoped I had made it clear in my introductory remarks that for the end of use sectors I had to make use of Table VI. These do not tie in precisely with the total Robinson forecast and I tried to interpret them to get a total as near the mean of Robinson as possible. Obviously the answer came out rather lower.

I am very surprised at Mr Jamieson's statement about nuclear energy, because I thought I had taken a figure right bang in the middle of the range, when one interpolates their figures for 1968. However, Mr Jamieson is a much better statistician and economist than I am, so I suppose I must accept that this figure is wrong.

I agree that the industry has got into a very tricky position and must make every effort to retain flexibility. Maybe we will be forced into a position which is not economically desirable or even viable. We must face the fact that owing to a series of circumstances in the last three or four years, partly due to the structure of the industry, we are in a position of extreme vulnerability. If we cannot hold our horses a bit we are going to come in for arbitrary restrictions. That is the point that we were trying to get over to the Arab Petroleum Congress in Beirut in October, that if prices go up too much, we naturally lose the marginal consumer, but that if prices are too low, we will create artificial restraint on sales. But in the present paper I did not try to tackle that problem; I wanted to see whether, from the Robinson Report, a change in the relative trends of different products was discernible. This report does, of course, give some warning on the coal/oil issue, and I took account of that in so far as I used the Robinson forecast for coal and assumed it would have to find an outlet. Obviously, any two people, working from such nebulous documents, would be bound to get different answers. But I would say this: that the one I worked out does seem to tie in with common sense, namely, a definite increase in the proportion of middle distillates, a decrease in the proportion of residuals, but a rather indeterminate change in the relative amount of gasolines. The rather laborious calculating simply put figures to what I felt was a natural consequence of the present position in Europe.

P. F. Miles (E. I. Du Pont de Nemours & Co.): May I raise a question not directly related to the subject of energy, but which may have a bearing on the question of the product-mix. This is the use of petroleum fractions as raw material for the chemical industry. In Europe the different fuel pattern compared with the U.S.A. has brought with it a considerable amount of subsidiary cracking of light naphtha fractions to produce olefins, and a demand for naphtha for this use has, therefore, been created and is likely to increase in the future. As natural gas becomes available in larger quantities in Europe, I assume increased consideration will be given to the use of this material not only for fuel but also as a raw material for chemical synthesis. It is appreciated that the actual amounts of naphtha and natural gas involved may be small compared with the fuel sector, but have they been considered in Mr Hubbard's analysis?

Although imported natural gas from overseas is likely to prove expensive, I have noted that a number of chemical companies in Italy are associated with pipeline import projects from North Africa, particularly for Sicily, situated close to the North African coast.

M. E. Hubbard: The Robinson Report did not touch on the petrochemical side, it dealt with energy; so I tried to leave it aside in these calculations, but I did say that one must take some account of the products and raw materials required for non-fuel purposes. Unfortunately the figures given in the OEEC statistics do look rather suspicious. They would seem to be on the low side. Petrochemicals have the great advantage of giving an added flexibility to the oil industry in producing its fuels, by choosing the part of the barrel out of which to make petrochemicals. It does not require a big amount that you can swing from right to left to make

a very material improvement in the flexibility of meeting market demand from one or from a series of refineries.

As Mr Miles says, petrochemicals represent only a small proportion of the total. It may reach, say, 2½ per cent of total oil demand, though it will be probably quite a long time before it reaches even that percentage. And total oils represent 30 per cent of total energy, and so it can be seen, for a long time ahead, that the influence of petrochemicals is unlikely to exceed 1 per cent of the total fuel and energy requirements.

C. I. Kelly: I am very interested in Mr Hubbard's figure, in Table VI, for the estimated consumption of natural gas in 1968. He has put it at a total of 48 million tons of coal equivalent (MTCE). This, he says in the body of the paper, is made up of three items: about 15 MTCE which he has assumed will be imported by pipeline into Europe, in 1968; 3 MTCE likely to enter Europe as liquefied natural gas in tankers; and finally he adds Robinson's 30 MTCE to cover production of indigenous gas. None of us has serious grounds for complaint against Robinson's 30 MTCE for indigenous gas, and I will align myself with Mr Hubbard on this quantity.

The 18 MTCE amount remains for us to argue about. Mr Hubbard is a better guesser than I. However, I wonder whether we ought to write off this amount altogether; that this may not enter Europe in 1968 is suggested by the rather impressive thesis propounded in Piacenza a few months ago by Dr Frankel. If I interpret him correctly, he thought that the introduction of natural gas into Europe would possibly be combated by the oil companies who could bring into the European energy market low-priced incremental crude oil production from oilfields abroad. Does Mr Hubbard think there is any substance in such an idea?

M. E. Hubbard: I do not think it is going to be handled that way. I think France wants to get that gas from the Sahara, and whether it is profitable on paper or not they will, I think, make every possible effort to lay a pipeline.

Some people say that the technical problems of crossing the Mediterranean are such that they will shy away. Well, the technical problems of the Lacq gas field were far greater, in several respects, than any that had ever been met before by the natural gas industry. Yet they were selling gas in five years. I think that if the French could tackle the Lacq gas field so remarkably effectively, they can most probably succeed with the problem of crossing the deep sea. Personally, but I would very much like to hear Mr Ornstein's views on this. I should put my money on there being a pipeline rather than against, and in a relatively short time.

Dr P. H. Frankel (Petroleum Economics Ltd): I feel I should add something to what Mr Kelly said. I am a little hesitant to do so, for he knows so much more than most of us about the question of natural gas and its handling and transport. But I believe he has also made a point which I tried to make elsewhere that we should not consider, as undoubtedly Mr Hubbard did to-night, in an almost Marxian way, the long-distance transport of natural gas as an historical inevitability. This, I think, one can only do if, as Mr Hubbard has already done on another occasion to-night, one puts political expediencies so very much higher than economics. In his paper, he does not deal at all with comparative prices and costs. I know the inhibitions in which people are involved even when they are retired from oil companies, and the handicaps and difficulties under which they operate—but I do not think it is possible to look at this statistical exercise of Mr Hubbard's entirely without numbers attached to it. And when I say numbers, I do not necessarily mean percentages.

But to go back to the question of natural gas to be transported over a long distance—say, at ultimate range, it is certainly possible technically to solve this problem. It is probably possible to solve many technical problems to-day which we are not called upon to solve economically. I feel that it would be necessary, before one accepts the estimates of people who want either to sell pipe or to sell gas, before one takes these estimates at their face value, for more work to be done and for more thinking to go into the relationship of the oil and gas industries.

We have a certain historical precedent in the U.S.A. I was present at the IP meeting seven years ago when Mr Hubbard read his original paper, and I believe it was my privilege to open the discussion on that occasion. At that time he certainly opened the eyes of the audience to the enormous importance of natural gas in the energy picture of the U.S.A. And that, in effect, the natural gas there did what fuel oil now does in Europe. But this position of co-existence, peaceful or otherwise, of natural gas and fuel oil in the U.S.A. has created a number of problems which deserve to be studied in Europe, because some of the problems may arise here, not in the same fashion, but problems of the same order. I believe Mr Kelly may have seen a simplified version of an already simplified paper which I read last autumn, and has not entirely brought out my point in saying that the oil companies may do better than they do now and sell fuel oil more cheaply. On that occasion, I only tried to point out that the so-called economics of transport of natural gas over very long distances and under difficult, *i.e.* costly, circumstances, that the economics at the receiving end depend on a very low valuation of natural gas at source. I have only suggested that we should learn from the U.S.A., which for too long, for various reasons, technical, commercial, and legal, has considered the natural gas and the oil industry as two separate industries, operating in self-contained spheres. If we do not make the same mistake in Europe we will see what might happen if we were to apply elsewhere the system which the proponents of some of the long-distance gas pipelines apparently have in mind, *i.e.* that one has found the gas anyhow, and that the natural resource has to be used, and used instantly, because it is a crime not to. If one were to apply the same system, the same way of thinking to the reserves in certain Middle East countries, which at the present rate of exhaustion would be called upon to be used a hundred years from now. If one were to apply the same principle towhat Mr Kelly called the incremental barrel in certain areas of crude production, if one were to apply the same system to crude oil, incremental crude oil, just as one applies it to the gas, which one considers to be incremental just because it happens to be in a different natural state, we would find that long-distance transport of gas is a non-starter, because it is very much more expensive to transport gas than crude oil. This is especially the case in a position of tanker surplus, which we have to-day, and which we are likely to have for some time to come.

To conclude, I would certainly say that we should widen the scope of Mr Hubbard's paper in two directions, both of them economic. We should look at the comparative price or cost of the various supplies of energy; we should look particularly at the economic relationship of the natural gas and the oil industry.

M. E. Hubbard: How I agree with you, Dr Frankel, on that last statement. Of course, the transport of natural gas is a lot more expensive than the cost of transport of liquid oil. And it would never have been used on such

a scale in the U.S.A. if it were not for the reasons that Dr Frankel gives; on the average, natural gas at source costs only about one-sixth of the price of crude oil. This is, of course, due to the much lower royalties on gas. One might almost say the oil industry in the U.S.A. is at a very unfair disadvantage. Dr Frankel also mentioned the cost of the incremental ton of crude oil. In the Middle East, the cost of the production is so small compared with the payments to the producing governments that the incremental ton cannot be much cheaper than the average for the total production. And from what one gathered in Beirut, payments to the owners of the mineral rights are not likely to be reduced.

When it comes to natural gas we have in mind the Sahara. Here we have a country completely different from those in the Middle East, with a different outlook, different interests, and closely integrated with Europe. If they want to get their gas into foreign trade they can well sell it as much cheaper than Middle East oil as American gas is cheaper than American oil. They can always arrange for the gas to show a profit at the consuming end if they have complete control over the charge made at the source. So I do not think that the strictly economic approach is, in fact, valid under present conditions.

Dr. P. H. Frankel: May I add two other remarks to the previous ones? Of course, Mr Hubbard is perfectly right in saying that the taxes or royalties, or the profit-sharing on oil in the producing countries, have reduced the cost advantage which these oils might have had earlier on. But, there again, we see this confusion. The same countries which insist on high profits on oil will be prepared to let the gas go at a very much lower rate of tax, because they still do not see that it is really one and the same problem. I think that what is really needed in our conditions to-day is a greater educational effort in the widest sense of the word, to make clear and press home the essential economic features of the facts.

Another point that Mr Hubbard made was that the French will insist on it. The surprising item is that the French quite openly say that they do not really want that Sahara gas themselves. They want to sell it to other European countries. I believe that there is a lot more to be studied on these matters than has been done hitherto.

I would like to deal in passing with the question about the use of non-European natural gas in Europe for petrochemicals. It is somewhat doubtful that long-range transport of gas would be cheap enough to compete with naphtha coming from European sources. The use of non-European gas will probably be in spheres other than petrochemicals—the reason why natural gas is used in the U.S.A. for the petrochemical industry and not naphtha, is that gasoline or petrol, *i.e.* naphtha, has a good sale in the U.S.A. compared with natural gas. In Europe it is the reverse.

P. F. Miles: I was interested in this matter because I have noticed that a number of chemical companies, in Italy for example, are associated with pipeline products; therefore these people are at least studying the feasibility of using imported, presumably North African, gas as a raw material for chemical synthesis—presumably in Sicily, which is the nearest point, but it is still part of Europe, and therefore will affect the European mix, which we are discussing.

C. I. Kelly: I would like to add one more statement to make it clear that when I used the phrase "incremental crude oil," I employed a phrase not of my own coining. To clear up the matter, I will read a short paragraph found in Dr Frankel's original 1960 Piacenza paper. It goes like this: "... If it is considered desirable, and politically feasible, to increase the sum total of petroleum hydrocarbon supplies to energy-importing areas, this could—to make an extreme case—be achieved more smoothly and more cheaply by earmarking a certain amount of incremental crude oil [his phrase, not mine] from exporting areas which are in a position and would wish to increase their output at prices arrived at by methods akin to those by which natural gas is sometimes found at source (see above, Section V). Then one would at least, more often than not, use the existing means of transport, especially tankers, of which there would appear to be a surplus supply for some time to come."

I will not elaborate more, now, for the subject could well become the title of a short dissertation, but I wish to state that in many of the crude oil exporting areas the relationship between the price of natural gas at source and that of the crude is fixed, and in them natural gas is abundantly available for exportation to energy-hungry markets.

But which crude oil exporting areas may wish to yield low-priced incremental crude oil for the suggested purpose, the author of the quoted paper did not make clear. Are they in North Africa, the Middle East, or would the oil be Russian oil at a dollar a barrel at Black Sea ports?

J. Ornstein: I personally believe that natural gas, if it is available in huge quantities, at about 400 miles from the inland sea of the Mediterranean, will be used in Europe. Whoever it belongs to, whether it is the French Government which "bends backwards" or anybody else, it will be used. And if the economics, which Dr Frankel mentioned, are far from being clear to-day, they will be clear one day and I believe within the years up to 1968.

I had the honour of sitting in innumerable meetings with the OEEC oil and gas committees, delving deep into the economics of gas production and gas transport, and I saw the efforts, as Dr Frankel puts it, of pipelayers, pipe producers, and gas consumers, to study this subject. From my experience, however, it appears that whatever we do, for or against it, whatever these incremental possibilities may be, I think the natural gas from the Sahara will be produced, transported, and brought somehow to Europe. Whether France will take its share, how much that share will be, and whether France will share the interest on the gas is in my opinion of no importance. The gas is there and will demand to be consumed, although it will probably not be consumed in such huge quantities as was expected originally. We have seen similar optimistic situations before, but it will come to the markets and I believe that Mr Hubbard is not too far off the mark with his figures.

If I may be allowed to put a question arising from the very interesting contribution by Mr Dalton; does he not think that the seasonal fluctuations of the future consumption of middle distillates, which he rightly fears will be strongly counterbalanced by the development of road and rail consumption of diesel oil, are increasing in the opposite direction, as in the consumption of diesel oil for domestic heating? I think that we as marketers over-estimate the influence of these seasonal fluctuations.

C. P. Dalton: Perhaps I did not make my point quite clear. The fluctuations I referred to concerned the demand for fuel oils and not for middle distillates. I had in mind that coal is relatively inelastic in supply and that the surges due, for example, to changes in economic activity and to temperature differences will mainly affect the demand for fuel oil.

A vote of thanks to the author was accorded with acclamation.

AUTOFINING STUDIES WITH MODEL COMPOUNDS *

By W. J. HENDRIKS †, J. van RYSSELBERGE ‡, J. C. VLUGTER † (Fellow), and H. I. WATERMAN † (Honorary Fellow)

SUMMARY

Autofining experiments with model compounds were performed in a flow reactor containing a fixed bed of cobalt oxide–molybdena–bauxite catalyst. Feedstocks were composed mostly of a hydrogen donor (decalin or tetralin), a sulphur compound (dibenzothiophene), and a diluent (benzene). In addition, a cycle oil fraction was desulphurized by an autofining operation, decalin being used as a hydrogen donor. In all the experiments a high degree of desulphurization was obtained.

In the paper detailed attention has been given to the transformation of the naphthenes.

It has been found that the isomerization of six-membered rings into five-membered rings has to be considered as an important side reaction in the conversion of the naphthenes. In view of the autofining process, this reaction results in a waste of part of the hydrogen which is potentially available in the six-membered ring naphthenes of the feedstock.

Autofining operating conditions are dictated by the type and the quantity of naphthenes present in the feedstock. If it is desired to eliminate appreciable amounts of sulphur from the feedstock at the higher pressures which will prevent rapid decline of catalyst activity, naphthene content of the feedstock should be high. In particular, the high pressure autofining of an aromatic oil fraction will be unfavourable with respect to the hydrogen balance. At the higher pressures the conversion of the naphthenes which have to be added to the feedstock will be relatively low. In addition, the aromatic rings of the compounds in the feedstock will in part be hydrogenated into naphthenic rings.

INTRODUCTION

AUTOFINING is one of the catalytic processes for the desulphurization of petroleum distillates.[1, 2] The most important reaction in these processes is the elimination of sulphur as hydrogen sulphide by means of catalytic hydrogenation of the sulphur compounds present in the feedstock. However, the essential feature of the autofining process is that the hydrogen required for this reaction is obtained by simultaneous dehydrogenation of six-membered ring naphthenes which are present in the processed distillate. In comparable hydrofining processes, *e.g.* the Unifining process, the Shell trickle process, the hydrogen required for the desulphurization reaction is supplied from another source, *e.g.* a Platforming unit. In practice, the difference between autofining and hydrofining is less sharp because the term autofining is sometimes still used if only a part of the required hydrogen is produced by simultaneous dehydrogenation of naphthenes.[3] In this paper the authors deal only with autofining experiments in which no hydrogen is supplied during the test runs. Extraneous hydrogen is used only in pressuring the unit.

In applying the autofining process some extra conditions have to be considered in comparison with normal hydrofining processes. In the first place the feedstock must contain sufficient naphthenes which can be converted into aromatics and hydrogen. If necessary a naphthene containing distillate can be added to the feed. Secondly, the processing conditions differ considerably from those used in hydrofining processes. Higher temperatures (400°–430° C) and lower pressures (5–15 atm) are required in order to promote the conversion of naphthenes into aromatics. This results in an increased deposition of coke on the catalyst, and consequently a more rapid decline of catalyst activity. Regeneration of the catalyst must be carried out rather frequently. Porter,[1] in processing middle distillates, aimed at a minimum on-stream time of 200 hours before regeneration of catalyst would be necessary.

The main reason for the present experiments with pure compounds was the lack of knowledge of the behaviour of naphthenes in desulphurization processes. It cannot be denied that, for instance in autofining middle distillates, it is more easy to follow accurately the desulphurization reaction by simply determining sulphur contents than it is to evaluate the conversions of the naphthenes. Despite the essential role of naphthenes in the autofining process, there is practically no published material available which deals with their conversions under autofining conditions. Szebényi [4] published in 1958 the results of batch autofining experiments involving the desulphurization of a gas oil fraction in the presence of pure naphthenes. The work described in this paper is concerned only with experiments in a flow reactor.

EXPERIMENTAL

Starting Materials

An evaluation of the starting materials is given in the dissertation from which the work described in this paper forms a part.[5] A short survey is given here.

* MS received 11 October 1960. † University of Delft, Holland. ‡ Labofina, Brussels.

Naphthenes. *Trans*-decalin, mixtures of *trans*- and *cis*-decalin, and tetralin were used as hydrogen donors. Purchased mixtures of the decalins contained about 80 per cent of the *trans* and about 20 per cent of the *cis* isomer and were used without purification. *Trans*-decalin was obtained by fractionation of such a mixture of decalins. Commercially available tetralin required purification by repeated washing with sulphuric acid, and was finally distilled from sodium.

Sulphur compounds. Dibenzothiophene or a sulphur-containing cycle oil fraction was used in making up a feedstock. Pure dibenzothiophene was prepared according to Gilman and Jacoby.[6] The cycle oil fraction was obtained in fractionating a Middle East light catalytically cracked cycle oil.

Solvent. Pure benzene was used as a diluent and a solvent in all experiments where a mixture of naphthene and dibenzothiophene was processed.

Hydrogen. Commercial hydrogen of purity >99·7 per cent vol has been used in pressuring the unit and for activation of the catalyst.

Catalyst

An industrial catalyst was used which consisted of a mixture of cobalt and molybdenum oxides on bauxite; the catalyst contained about 8·5 per cent wt MoO_3 and about 1 per cent wt CoO. It is normally used for the hydrodesulphurization of oil fractions. The available catalyst particles were powdered, mixed with 1 per cent wt of graphite, and formed into cylindrical pellets (dia = 3 mm, height = 2·5–3 mm). These pellets were calcined at 550° C. Duration of the air treatment was 3 hours for the first batch of catalyst (*cf* Experiments, Table I). Because in the meantime it had appeared that a more active catalyst was produced by a prolongation of the calcination period, all further batches of catalyst were calcined for 10–20 hours.

TABLE I

Constant Pressure Autofining of a Synthetic Feed which is Composed of Decalin, Dibenzothiophene, and Benzene

Cat. No. CoMo-HD7

Composition of feed	Dibenzothiophene: 5·0 mol % Decalin*: 15·0 mol % Benzene: 80·0 mol %	sulphur content 1·74% wt		
Composition of recycle gas	Hydrogen sulphide: 2·3–2·7% vol Light hydrocarbons: 1 – 1·5% vol Hydrogen: 96 –96·5% vol			
Conditions of reaction	Temperature: 415° C Pressure: 13·0 atm abs Space velocity: 11·0 mol liquid feed/(l cat) (hr) Recycle rate: 2·8 mol gas/mol liquid feed			
Test period no. Hours on stream		1 10–16	2 16–22	3 22–28
Conversion (%) of decalin into	Methylindanes	19·6	18·4	18·2
	Tetralin	11·7	11·7	10·4
	Naphthalene	42·2	42·6	42·1
	Mono-aromatics, total	31·3	30·1	28·6
	Aromatics, total	73·5	72·7	70·7
Conversion (%) of dibenzothiophene into	Biphenyl	91·2	90·0	87·8
Hydrogen balance (mol H_2/mol decalin)	Net over-production (measured)	2·24	2·17	2·04
	Consumption (calculated from dibenzothiophene conversion)	0·61	0·60	0·59
	Totally formed by dehydrogenation of decalin	2·85	2·77	2·63
	Hydrogen formation corresponding with measured conversion of decalin into:			
	Mono-aromatics	0·94	0·90	0·86
	Naphthalene	2·11	2·13	2·10
	Total aromatics	3·05	3·03	2·96

* 79·6% *trans*-decalin and 20·4% *cis*-decalin.

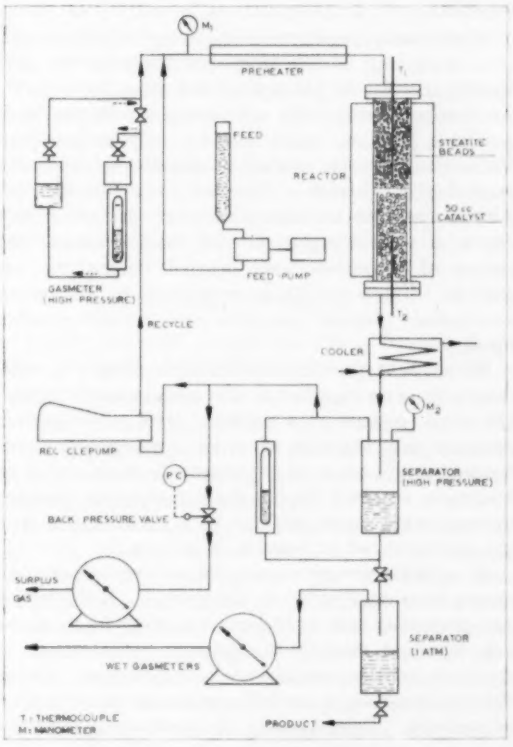

FIG 1

DIAGRAM OF BENCH SCALE UNIT FOR AUTOFINING EXPERIMENTS

Finally, the calcined catalyst was activated with hydrogen. Hydrogen activation was carried out in the reactor at 425° C and 15 atm and lasted 3 hours.

Apparatus

In Fig 1 a diagram of the flow system is given. The essential part of the apparatus is a stainless steel high pressure flow reactor. The lower part of the reactor contains a 50-cc fixed bed of catalyst pellets, while the upper part is filled with steatite beads and functions only as a preheating section.

The back pressure valve is in operation only when the unit is operated at constant pressure and excess hydrogen is produced by dehydrogenation of the naphthenes. In this case the high pressure gas (HP

gas) which leaves the high pressure separator is partly recycled and partly vented through the back pressure valve. The liquid product in the high pressure separator always contains some dissolved gas, mainly hydrogen, which is set free as the product passes into the low pressure separator. This gas is designated low pressure gas (LP gas).

For special features of the apparatus, *e.g.* the high pressure gas meter, one is referred to the literature.[7]

Procedure

When catalyst activation was completed, the hydrogen circulating in the unit was replaced by fresh hydrogen. In the pre-test period subsequent to the catalyst activation, the processing conditions (temperature, pressure, space velocity, recycle rate) were first adjusted to the desired level and then kept constant during the rest of this period. During the test runs temperature and space velocity were always kept constant. With regard to the total pressure, two modes of operation were applied during the test periods. These operations are respectively designated as constant pressure operation and variable pressure operation.

In constant pressure operation, hydrogen is withdrawn from or supplied to the unit in order to keep the total pressure at a constant level. The authors consider only the case when the hydrogen produced by dehydrogenation of naphthenes is in excess of the hydrogen required for the desulphurization reaction. Consequently, hydrogen has to be withdrawn from the unit to avoid an increase of pressure.

In variable pressure operation no hydrogen is withdrawn from or supplied to the system. If the hydrogen-producing and hydrogen-consuming reactions are not balanced exactly the pressure will increase or decrease until an equilibrium is established. Excess dehydrogenation of naphthenes results in an increase of pressure. Consequently, the pressure is lowered if hydrogen-consuming reactions are dominating. As a consequence of pressure changes, the recycle rate will change accordingly because the recycle pump is operated at a constant volume capacity.

It is noted that in both operations a relatively small amount of hydrogen is always withdrawn from the system. This is due to the solubility of hydrogen in the liquid product in the high pressure separator (*cf* Fig 1).

METHODS OF ANALYSIS

Gases

The composition of all gas streams was determined at regular intervals. Hydrogen sulphide content was determined volumetrically. After the removal of hydrogen sulphide by washing with alkali a GLC analysis was made. Olefinic hydrocarbons could never be detected. The HP gas always contained minor amounts of light hydrocarbons, predominantly methane. The concentration of light hydrocarbons in the LP gas was sometimes high. In processing mixtures of decalin and cycle oil, this gas contained up to 20 per cent vol of light hydrocarbons.

Liquids

In autofining a feedstock which contains dibenzothiophene and one of the hydronaphthalenes, the following hydrocarbons can be expected to be present in the reaction product: *trans*-decalin, *cis*-decalin, tetralin, naphthalene, biphenyl, *cyclo*hexylbenzene, and bi*cyclo*hexyl. Mixtures of these compounds can be analysed rapidly by GLC.[8] In practice, however, it appears that more complicated reaction products are formed, especially when the decalins are used as hydrogen donors. The composition of the product obtained when processing a feedstock consisting solely of *trans*- and *cis*-decalin is therefore discussed in detail. A sample of the crude product was dissolved in benzene and the solution brought into the GLC analyser. The chromatogram is shown in Fig 2 (*a*). It is immediately apparent that some other compounds besides tetralin are present in the tetralin fraction. A concentrate of these compounds was prepared by fractionating a sample of the crude product in a spinning band column. A GLC analysis of the concentrate is shown in Fig 2 (*b*). Besides tetralin, two distinct compounds, designated as x_1 and x_2, are present.

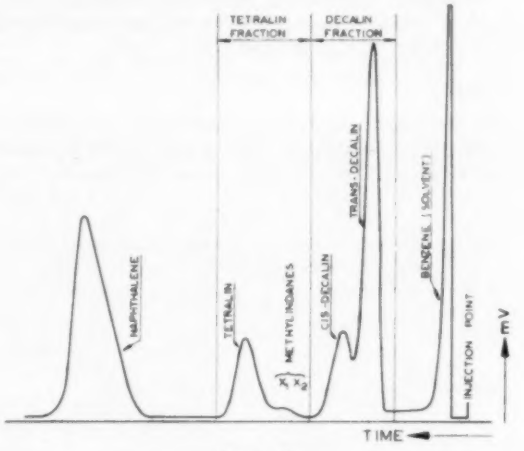

FIG 2 (*a*)

CHROMATOGRAM OF A CRUDE PRODUCT OBTAINED IN PROCESSING DECALIN OVER DESULPHURIZATION CATALYST

The compounds x_1 and x_2 were of an aromatic nature, as was shown by FIA analysis. An infra-red analysis gave evidence for the presence of 5-methylindane. It seems reasonable to consider respectively x_1 and x_2 as 4- and 5-methylindane (boiling points of 5-methylindane, 4-methylindane, and tetralin are re-

spectively 201·8° C, 204·4° C, and 207·6° C). It must be noted that in the meantime Hastings and Nicholson have published infra-red data of all the methylindanes.[9]

Another complication was observed in the decalin fraction of the product. This fraction was isolated from a sample of the crude reaction product by fractional distillation. On GLC analysis the isolated fraction showed peaks only in the decalin region. However, the fraction was not solely composed of decalins. A FIA analysis showed that an important quantity of aromatics was also present in the fraction. These aromatics were isolated from the fraction by percolation in a silica gel column[10] and the mixture of aromatics investigated by GLC analysis. At least two aromatic compounds were present. The chromatogram showed one peak representing a compound with the same retention time as *trans*-decalin and a predominating peak representing a compound with the same

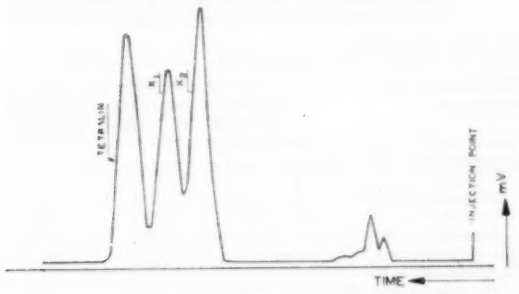

FIG 2 (*b*)

CHROMATOGRAM OF A DISTILLATE FRACTION OBTAINED FROM THE PRODUCT SHOWN IN FIG 2 (*a*)

retention time as *cis*-decalin. Chromatograms of standard mixtures of pure compounds showed that 1- and 2-methylindane had practically the same retention time as *cis*-decalin and that indane and butylbenzene had practically the same retention time as *trans*-decalin. An infra-red analysis of the aromatics, isolated from the decalin fraction of a crude product, gave evidence for the presence of methylindanes and butylbenzene.

After the investigation of a large number of products obtained in dehydrogenation experiments with the decalins and in autofining experiments with the decalins as hydrogen donors, the authors are convinced that methylindanes, indane, butylbenzene, and perhaps small amounts of other C_{10} alkylbenzenes are formed whenever a cobalt oxide–molybdena–bauxite catalyst is used. It is believed that the methylindanes will predominate in this series of side products when autofining conditions are used. Therefore the complex mixture of the said aromatics will simply be designated as methylindanes. Whenever the designations " decalin fraction " and " tetralin fraction " are used they will have the same meaning as in Fig 2 (*a*).

The hydrogenation products of dibenzothiophene caused less analytical difficulties. This sulphur compound was rather selectively converted into biphenyl, whereas hydrobiphenyls were nearly absent. The amount of unconverted dibenzothiophene was calculated from sulphur determinations. Sulphur contents were determined by the quartz tube method.[11]

Olefinic products, *e.g.* methylindenes and dihydronaphthalenes, could not be detected. Any appreciable amount of unsaturates would have been found by an ozonolysis or an analytical hydrogenation.[12] These methods were applied on several distillate fractions, as well as on crude products.

In analysing an autofined cycle oil fraction, statistical methods were used, for which one is referred to the literature.[13]

Analysis of autofined products was generally made after the removal of hydrogen sulphide by washing with a sodium hydroxide solution and subsequent drying with calcium chloride or magnesium sulphate.

PROCESSING RESULTS

1. *Constant Pressure Autofining with a Benzene Solution of Decalin and Dibenzothiophene as a Feedstock*

Table I summarizes the results of three successive test runs, each of 6 hours duration, which succeeded a 10-hour pre-test period. In considering the conversions, it is found that about 70 per cent of the decalins was dehydrogenated to aromatics.

With time on stream a slight decrease of the conversion into mono-aromatics (tetralin, methylindanes) was observed. It proves that the conversion into methylindanes was almost twice as high as the conversion into tetralin. Obviously the methylindanes are undesired side products. In the autofining process the isomerization to five-membered rings must be interpreted as a waste of hydrogen which is potentially available from the six-membered ring naphthenes of the feedstock. Hitherto the importance of this reaction has been neglected in the autofining literature.

About 90 per cent of the dibenzothiophene was converted into biphenyl. The conversion slightly decreased with time on stream. The conversion of dibenzothiophene as calculated from sulphur determinations was in accordance with the conversion calculated from biphenyl recovery. This means that the sulphur compound was rather selectively hydrodesulphurized to biphenyl. The benzene content of the autofined solutions amounted to 69·2–69·5 per cent wt. This is only slightly more than the theoretical benzene content (69·0 per cent wt), which has been calculated on the supposition that the weight ratio (benzene)/(dissolved material) had changed only because of the conversion of dibenzothiophene into biphenyl and of the conversion of decalin into C_{10}-aromatics as shown in the table. Moreover, the actual

recovery of dissolved material amounted to 97–97·5 per cent of the theoretical recovery which should have been obtained if the said supposition was right. These values indicate that during the test runs small amounts of dissolved material were converted into catalyst deposit and probably into benzene by destructive hydrogenation.

Finally, a hydrogen balance is given in Table I. The excess hydrogen was calculated from composition and quantity of surplus gas (*cf* Fig 1). It appears that more than 2 mol of hydrogen/mol of processed decalin had to be withdrawn from the system in order to avoid an increase of pressure. The hydrogenation of dibenzothiophene into biphenyl theoretically demands a production of 0·6 mol/mol of processed decalin. In this way, it is estimated that the total hydrogen production for the first test run was 2·85 mol/mol of decalin (*cf* Table I). The hydrogen production theoretically corresponding with the measured conversions of decalin into aromatics is slightly higher (3·05 mol/mol of decalin). The difference is explained in part by the loss of the hydrogen which escaped in the LP gas and probably in part by hydrogen-consuming side reactions which may have occurred to a small extent (destructive hydrogenation, saturation of aromatics).

Comparative Dehydrogenation Experiments

The same batch of catalyst which was used in the autofining experiments described in the preceding section was employed in comparative dehydrogenation experiments with a feedstock solely composed of decalin and benzene. A dehydrogenation run was made before and after the autofining runs. Table II summarizes the operations with this batch of catalyst.

The catalyst has not been in operation continuously during the whole set of experiments. After dehydrogenation experiment I, as well as after autofining experiments II, the catalyst was cooled down to room temperature in a stream of hydrogen and stored in the reactor under hydrogen pressure (15 atm). Before the start of each of the three operations the catalyst was treated with hydrogen for 3 hours at the usual activating conditions (425° C, 15 atm, 1000 l H_2 NTP/(1 cat) (hr)). After the last dehydrogenation experiment the catalyst was finally purged with nitrogen for one hour (400° C, 85 atm, 5000 l N_2 NTP/(1 cat) (hr)). It appeared that the spent catalyst contained 3·5 per cent of sulphur and 6·0 per cent of carbonaceous material.

Table II gives the distribution of the C_{10} hydrocarbons in the products from the three operations. It is immediately seen that the conversions of decalin differ remarkably for the three sets of experiments. These differences are observed notwithstanding the identical decalin content of the processed feedstocks. In comparing the results of the successive operations it must be noted that a large difference was expected between the conversions obtained in dehydrogenation runs I and III. The relatively small conversion of decalin in the last dehydrogenation run was considered not surprising because of the sulphiding of the catalyst and the deposition of carbonaceous material on the catalyst, both of which had occurred during the processing of the sulphur-containing feed in the preceding autofining operation.

Hitherto, sulphiding of the type of catalyst employed was considered detrimental to its dehydrogenation activity because of the authors' experience

TABLE II

Comparison of Conversions of Decalin Obtained in Respectively Processing a Benzene–Decalin Feed and a Benzene–Decalin–Dibenzothiophene Feed over Co–Mo–Al_2O_3 Catalyst

Conditions of reaction:
Temperature: 415° C
Pressure: 13 atm abs
Space velocity: 11 mol liquid feed/(1 cat)(hr)
Recycle rate: 2·8 mol gas/mol liquid feed

Cat. No. CoMo-HD7

Experiment no.	I	II	III
Operation	Dehydrogenation	Autofining (*cf* Table I)	Dehydrogenation
Composition of feed (mol %):			
Dibenzothiophene	0	5	0
Decalin*	15	15	15
Benzene	85	80	85
Duration of runs:			
Pre-test period, hr	3	10	3
Test period, hr	2	18	2
Total, hr	5	28	5
Composition of the C_{10}-hydrocarbon fraction in liquid products obtained during test periods (mol %):			
Decalin fraction	59	38–42	82
Tetralin fraction	17	16–19	8·5
Naphthalene	24	42	9·5
Hydrogen production, mol H_2/mol decalin	2·15	2·0–2·2†	1·0

* 79·6% *trans*-decalin and 20·4% *cis*-decalin.
† Over-production.

with pre-sulphided catalysts in the processing of pure decalin. With a pre-sulphided cobalt oxide–molybdena–bauxite catalyst the authors always obtained less conversion of decalin into aromatics than with the same catalyst in its oxide form. However, the relatively high conversions of decalin into aromatics observed in the autofining test runs (*cf* Table II) seem to contradict this experience. It is noticed that the excess hydrogen produced in the autofining runs equals the total hydrogen production of the first dehydrogenation experiment. In the autofining runs the hydrogen which was produced in addition to the

hydrogen withdrawn from the system (*cf* Table I) was consumed by the desulphurization reaction. Two possible causes which may be responsible for the relatively high conversion of decalin during the autofining test runs have to be considered. First, the whole history of the catalyst up to the first autofining test run can be considered as a special and effective pre-treatment of the catalyst. If decalin dehydrogenation is then considered as an independent reaction in the autofining operation the results of the last dehydrogenation experiment (III) must be disregarded. The low conversion of decalin in this last experiment must then be explained by unfavourable changes in catalyst structure and/or nature of deposits during the period between operations II and III.

A second possibility, however, is that the presence of the dibenzothiophene in the autofining feedstock promotes the dehydrogenation of decalin, not only in view of thermodynamic equilibria but also in a fundamental way. For the moment one cannot exclude, for example, a hydrogen transfer reaction.

2. *Variable Pressure Autofining with a Benzene Solution of Dibenzothiophene and Decalin or Tetralin as a Feedstock*

The composition of the feedstocks and the processing conditions are given in Table III. The feedstocks

TABLE III

Conditions of Reaction used in the Variable Pressure Autofining of Mixtures of Dibenzothiophene, a Naphthene, and Benzene

Catalyst no.	CoMo-HD8	CoMo-HD9	CoMo-HD10
Compounds in feed:			
Sulphur compound	Dibenzothiophene	Dibenzothiophene	Dibenzothiophene
Naphthene	Decalin*	*trans*-Decalin	Tetralin
Diluent	Benzene	Benzene	Benzene
Composition of feed: 5·0 mol % dibenzothiophene, 5·0 mol % naphthene, 90·0 mol % benzene } sulphur content = 1·86% wt			
Conditions of reaction:			
Temperature:	415° C		
Pressure:	is kept constant during the pre-test period (0–5 hr). After this pre-test period the pressure changes with time on stream. The observed pressures are given in Fig 3		
Space velocity:	13·5 mol liquid feed/(1 cat)(hr)		
Recycle rate:	linearly changes with total pressure (P atm abs). For all experiments the recycle rate at any moment is equal to: (0·15 × P) mol gas/mol liquid feed		

* 78·4% *trans*-decalin and 21·6% *cis*-decalin.

differ only with respect to the nature of the naphthenic compound. In order to avoid a large overproduction of hydrogen, the naphthene content of the feedstock was fixed appreciably lower than in the foregoing autofining experiments.

The total pressure of the unit was kept constant during the pre-test period. After the pre-test period it depends mainly on the conversions whether the pressure will change or not. Fig 3 shows the variation of pressure with time on stream. In processing the feedstock which contained both decalin isomers the pressure slowly increased. Average rate of increase in the period from 5 to 20 hours time on stream was 0·3 atm/hr. In processing the feedstock which contained only *trans*-decalin as a hydrogen donor the pressure remained practically constant. Evidently hydrogen production/mol of naphthene is higher if a mixture of the decalins is used instead of *trans*-decalin. The *trans*-decalin is obviously less reactive than its *cis* isomer. In separate dehydrogenation experiments with feedstocks consisting solely of benzene and one of the decalins, it has also been observed that the *cis*-decalin is more easily converted into aromatics than the *trans*-decalin.

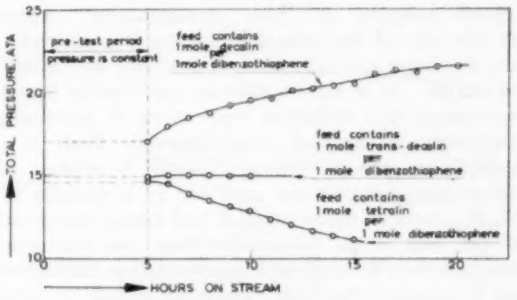

FIG 3

OBSERVED PRESSURES DURING THE VARIABLE PRESSURE AUTOFINING OF MIXTURES OF BENZENE, DIBENZOTHIOPHENE, AND A NAPHTHENE (*cf* TABLE III)

During the experiment with the tetralin-containing feedstock a steady decrease of pressure was observed. However, it must be noted that in this case a decrease of pressure could not be prevented, even if the conversion of tetralin into naphthalene had been complete. This appears from the theoretically possible conversions and the quantity of hydrogen which is lost by the removal of gas from the low pressure separator (*cf* Fig 1). This quantity of hydrogen amounts to only 0·005–0·010 mol/mol of processed feed but nevertheless becomes appreciable if related to the processed naphthene. The loss equals 0·1–0·2 mol of hydrogen/mol of processed naphthene. This means that for the operation with tetralin as a hydrogen donor, 10–20 per cent of the theoretically possible formation of hydrogen is lost merely by the withdrawal of the low pressure gas from the system.

Dibenzothiophene was desulphurized by more than 99 per cent in the three autofining experiments. Biphenyl was the predominant reaction product, whereas only relatively small quantities of the hydrobiphenyls were formed.

The minor saturation of the aromatic rings of the

sulphur compound can be considered as an advantage of autofining processing conditions. In a separate experiment the authors processed a benzene solution of dibenzothiophene (5 mol per cent) over a fresh batch of catalyst using operating conditions similar to those of the autofining process. At 415° C and 20 atm the solution, together with hydrogen (3·25 mol H_2/mol solution), was passed over the catalyst (13·5 mol solution/(1 cat) (hr)). Desulphurization was almost complete in these circumstances. About 93 per cent of the dibenzothiophene was converted into biphenyl and only about 7 per cent into hydrobiphenyls. It is known that an appreciably higher conversion into saturates may occur if processing conditions are applied which resemble those of a hydrofining process. Hoog [14] passed a solution of dibenzothiophene (8 per cent wt) in a paraffin oil ($C_{12}H_{26}$) over a similar catalyst and found that about 35 per cent of the dibenzothiophene was converted into *cyclo*hexylbenzene and bi*cyclo*hexyl at the following processing conditions: temperature 375° C, pressure 50 atm, space velocity 3 kg liquid feed/(1 cat) (hr), hydrogen rate 1000 l H_2 NTP/kg liquid feed. Only about 65 per cent of the dibenzothiophene was converted into biphenyl. However, in modern refining the saturation of aromatics in a hydrodesulphurization process is in many cases of little importance. Hydrodesulphurization of a light oil fraction is often followed by Platforming of the product, when most of the naphthenes formed by the saturation of aromatics in the first operation may in turn be dehydrogenated to aromatics in the second process.

TABLE IV

Conversions Obtained in the Variable Pressure Autofining of a Solution of Decalin and Dibenzothiophene in Benzene* (cf *Table III*)

Cat. No. CoMo-HD8

Test period no.	1	2	3
Hours on stream	5–10	10–15	15–20
Pressure, atm abs (*cf* Table III):			
At the beginning of the test period	17·0	19·5	20·5
At the end of the test period	19·5	20·5	21·6
Conversion of decalin into aromatics, %	94·6	94·5	96·5
Conversion of dibenzothiophene into hydrocarbons, %	99·6	99·4	99·3
Distribution C_{10}-hydrocarbons in product, mol:			
Decalin	5·4	5·5	3·5
Mono-aromatics (tetralin, methylindanes)	34·3	36·0	35·8
Naphthalene	60·3	58·5	60·7
Distribution C_{12}-hydrocarbons in product, mol:			
Hydrobiphenyl compounds (*cyclo*hexylbenzene, bi*cyclo*hexyl)	1·0	—	2·0
Biphenyl	99·0	—	98·0

* 78·4% *trans*-decalin and 21·6% *cis*-decalin.

In Table IV are summarized the results of the autofining runs with decalin as a hydrogen donor. The conversions were practically the same for the three successive runs. About 95 per cent of the decalin was converted into aromatics, 60 per cent into naphthalene, and about 35 per cent into mono-aromatics (tetralin and methylindanes).

Finally, the results of the autofining runs with tetralin as a hydrogen donor are given in Table V.

TABLE V

Conversions Obtained in the Variable Pressure Autofining of a Solution of Tetralin and Dibenzothiophene in Benzene (cf *Table III*)

Cat. No. CoMo-HD10

Test period no.	1	2
Hours on stream	5–10	10–15
Pressure, atm abs (*cf* Table III):		
At the beginning of the test period	14·6	12·9
At the end of the test period	12·9	11·2
Conversion of tetralin into naphthalene, %	83·1	90·0
Conversion of dibenzothiophene into hydrocarbons, %	99·5	99·0
Distribution of C_{10}-hydrocarbons in product, mol %:		
Decalin	0·7	0·0
Mono-aromatics (tetralin, methylindanes)	16·2	10·0
Naphthalene	83·1	90·0
Distribution of C_{12}-hydrocarbons in product, mol %:		
Hydrobiphenyl compounds	1·3	1·6
Biphenyl	98·7	98·4

Very little, if any, tetralin was converted into decalin. This shows that the conversion into naphthalene increased with increasing time on stream, probably because the dehydrogenation reaction was promoted by the decrease of pressure.

3. *Variable Pressure Autofining of a Cycle Oil with Decalin as a Hydrogen Donor*

In case an oil fraction which does not contain six-membered ring naphthenes has to be autofined, a naphthene or naphthene-containing oil fraction can be added to the feedstock. As an example the authors have processed a cycle oil fraction which contained very little or no compounds with naphthenic rings. Cycle oil is a middle distillate fraction obtained from catalytically cracked products. To this fraction decalin was added as a hydrogen donor. The composition of the two mixtures prepared and the processing conditions applied in the autofining of these mixtures are given in Table VI. The observed pressures are shown in Fig 4.

Pressure changed only slightly during the autofining of the mixture which contained 33·3 per cent wt

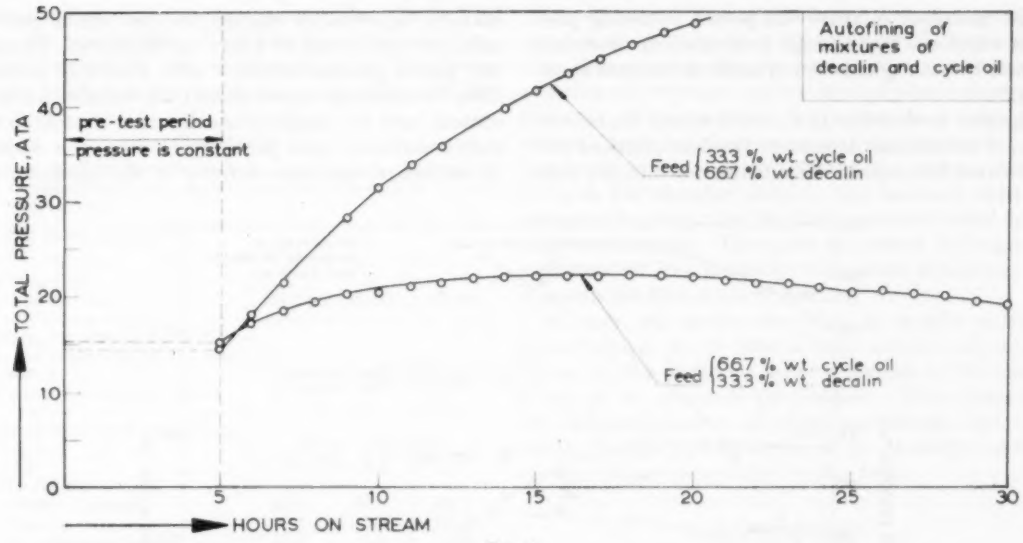

FIG 4

OBSERVED PRESSURES DURING THE VARIABLE PRESSURE AUTOFINING OF DECALIN–CYCLE OIL MIXTURES (*cf* TABLE VI)

of decalin. After the pre-test period, pressure slowly increased, until at 15 hours time on stream a pressure of 22·2 atm was achieved. In the period from 15 until 20 hours on stream, time pressure remained nearly constant, and then slowly decreased. After 30 hours on stream time, pressure was 19·3 atm, 4 atm higher than the pressure at the start of the experiment.

A rapid increase of pressure was observed in the processing of the feedstock, which was composed of two parts decalin and one part cycle oil. For the three successive periods 5–10, 10–15, and 15–20 hours time on stream, the average rate of increase respectively was 3·4, 2·0, and 1·4 atm/hr. After the pre-test period, pressure increased in 15 hours from 15 to 50 atm.

TABLE VI

Conditions of Reaction used in Autofining Mixtures of Decalin and a Cycle Oil Fraction

Catalyst no.	CoMo-HD11	CoMo-HD12
Composition of feed, % wt:		
Cycle oil fraction*	66·7	33·3
Decalin†	33·3	66·7
Sulphur content of mixture, % wt	2·05	1·03

Conditions of reaction:
Temperature: 415° C
Pressure: is kept constant during pre-test period (0–5 hr). After this period the pressure changes with time on stream. The observed pressures are given in Fig 4
Space velocity: 1·0 l liquid feed/(l cat) (hr)
Recycle rate: linearly changes with total pressure (P atm abs). The recycle rate at any moment is equal to: (66·5 × P) l NTP/l liquid feed

* Boiling range 290°–310° C/760 mm Hg; $n_D^{20} = 1{\cdot}5468$; $d_4^{20} = 0{\cdot}9457$; $M = 205$; sulphur content = 3·07% wt; $R_A = 1{\cdot}40$; $R_T = 1{\cdot}32$.
† 79·6% *trans*-decalin and 20·4% *cis*-decalin.

In these experiments the volume of gas removed from the low pressure separator (*cf* Fig 1) equalled, after the removal of hydrogen sulphide, 3–5 l NTP/l liquid product. This gas always contained more than 80 per cent vol of hydrogen, the balance being light hydrocarbons.

In all test runs the recovery of liquid product was higher than 96 per cent wt of the processed decalin–cycle oil mixture.

Fig 5 shows the desulphurization in relation to time on stream and pressure.

Relatively low operating pressures, as achieved in the processing of the feedstock with lowest decalin content, most probably result in a rather rapid decline of catalyst activity. The percentage sulphur removal decreased from 92 per cent to about 80 per cent during the on-stream period from 5 to 30 hours. In processing the feedstock which contained 66·7 per cent wt of decalin a rapid increase of both operating pressure and percentage sulphur removal was observed with increasing time on stream. Desulphurization was almost complete (>99 per cent) at the higher pressures.

These experiments reveal one of the economic problems of the autofining process. On the one hand, it would be attractive to add only a small quantity of naphthenes to the oil which has to be autofined. On the other hand, only a high percentage of naphthenes

in the processed mixture will permit operating pressures which are high enough to secure long on-stream periods before regeneration of catalyst becomes necessary.

In order to elucidate to a certain extent the conversions of decalin into aromatics, the desulphurized products were first separated by distillation into two main fractions. The quantities of light (<180° C) and heavy ends (>320° C) were small (*cf* Table VII). The boiling range of the two main fractions were respectively 180°–225° C and 225°–320° C. The first fraction, which contains decalin and its dehydrogenation products, was further investigated by GLC analysis. It proved that, with the aid of the chromatograms, this fraction could be divided into a decalin fraction, a tetralin fraction, and a naphthalene fraction. This sub-division could be made in an analogous way, as shown in Fig 2 (*a*), for a crude product obtained in the processing of pure decalin.

The tetralin fraction and the naphthalene fraction will certainly contain some hydrocarbons which have been formed from the cycle oil. Nevertheless, the tetralin fraction content and the naphthalene fraction content of the desulphurized products as calculated from the chromatograms will give an indication of the conversions of decalin. Figs 6 (*a*) and (*b*) show these contents in the desulphurized products for the two autofining experiments in relation to time on stream and pressure.

In autofining the mixture which contained 33·3 per cent wt of decalin, the composition of the product changed appreciably during the on-stream period only between 5 and 15 hours (*cf* Fig 6 (*a*)). During this period pressure changes were relatively strong. After 15 hours on-stream period the tetralin fraction content and the naphthalene fraction content both slowly declined, most probably because of a decline in the dehydrogenation activity of the catalyst. It has already been stressed that in this experiment the percentage sulphur removal decreased rapidly with increasing time on stream (*cf* Fig 5).

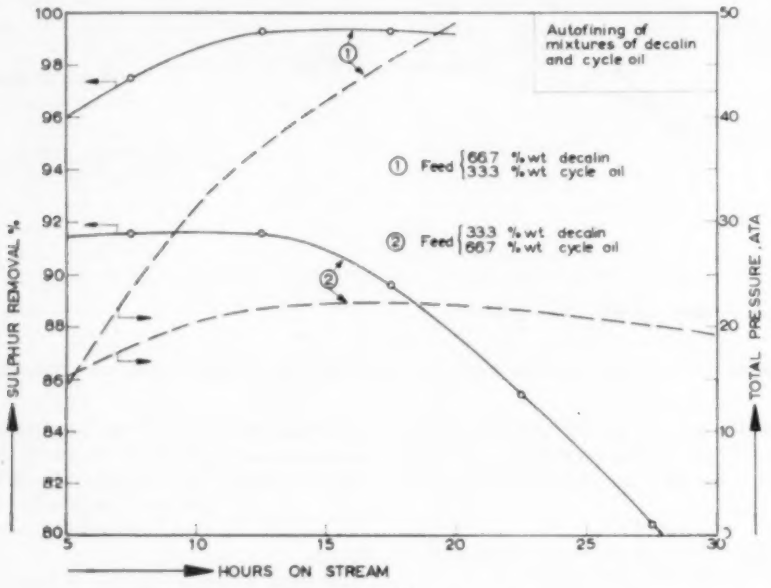

FIG 5

PERCENTAGE SULPHUR REMOVAL IN RELATION TO TIME ON STREAM AND PRESSURE (*cf* FIG 4)

The effect of the pressure is more evident for the autofining experiment with a feedstock containing 66·7 per cent wt of decalin (*cf* Fig 6 (*b*)). The large increase of pressure resulted in a marked increase of tetralin content and a corresponding decrease of naphthalene content. The same effect had already been observed in separate dehydrogenation experiments with pure decalin. The ratio (tetralin)/(naphthalene) in the products depended on hydrogen pressure as predicted by thermodynamics for the dehydrogenation equilibrium between tetralin and naphthalene.

It thus appears that a change of pressure during the autofining of a decalin–cycle oil mixture not only affects the conversions of decalin into tetralin and naphthalene, but in addition the composition of the desulphurized cycle oil also changes appreciably with pressure. The oil fraction of the autofined mixture (*cf* Table VII) will contain the hydrocarbons which were already present in the original cycle oil fraction, together with most of the hydrocarbons formed by

hydrogenation of the sulphur compounds. Table VII gives the composition of the fractions from the products obtained in the experiment with the higher decalin content in the feedstock. Comparison of the three successive test runs shows that the average number of aromatic rings per molecule (R_A) considerably decreases as the operating pressure increases, and that the average number of naphthenic rings per molecule (R_N) correspondingly increases. As the original cycle oil fraction contained practically no compounds with naphthenic rings, the results shown in Table VII therefore indicate that aromatic rings of compounds present in the feed were converted into naphthenic rings. The most important hydrogenation reaction was probably the partial saturation of di-aromatics into mono-aromatics.

Anyway, the results show that to operate at high pressures, *e.g.* 40–50 atm, a large addition of naphthenes to the feedstock will be needed in the autofining of an aromatic oil fraction. The relatively low dehydrogenation of these naphthenes and the relatively high hydrogenation of the aromatics of the oil fraction are responsible for this fact.

TABLE VII

Analysis of Products Obtained in Autofining a Mixture of Decalin (66·7% wt) and a Cycle Oil Fraction (33·3% wt) (cf Table VI)

Cat. No. CoMo-HD12

Test period no.	1	2	3
Hours on stream	5–10	10–15	15–20
Pressure, atm abs (*cf* Table VI):			
At the beginning of the test period	14·6	31·6	41·7
At the end of the test period	31·6	41·7	48·8
Distillate fractions* of product, wt %:			
<180° C	2·5	4·3	2·5
180°–225° C†—			
Decalin fraction	45·9	46·1	49·3
Tetralin fraction	9·5	13·5	15·5
Naphthalene fraction	15·6	9·0	6·3
225°–320° C oil fraction	25·8	26·7	26·1
>320° C residue	0·7	0·4	0·3
n–d–M analysis of oil fractions:			
n_D^{20}	1·5451	1·5301	1·5230
d_4^{20}	0·9358	0·9208	0·9139
M	200	200	200
Sulphur content, % wt	0·04	0·01	0·01
R_A	1·44	1·19	1·07
R_N	0·09	0·39	0·54
R_T	1·53	1·58	1·61

* Reported boiling ranges hold for a pressure of 1 atm.

† The sub-division of this fraction in a decalin, tetralin, and naphthalene fraction was possible with the aid of a GLC-analysis (*cf* also Fig 2 (*a*)).

CONCLUSIONS

The cobalt oxide–molybdena–bauxite catalyst investigated is active for both the hydrodesulphurization of sulphur compounds and the dehydrogenation of six-membered ring naphthenes into aromatics. An important side reaction in the conversion of the naphthenes is the isomerization of six-membered rings into five-membered rings. The formation of methylindanes from decalin is mentioned as an example. In the autofining process this isomerization reaction can be considered as a waste of part of the hydrogen which is potentially available in the feedstock.

Autofining operating conditions are almost entirely

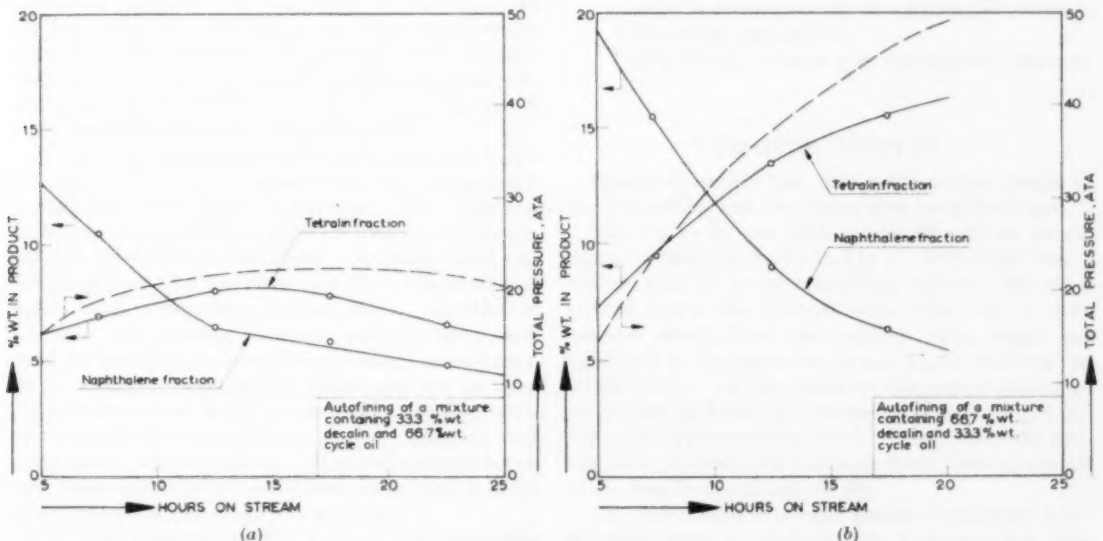
% WT IN PRODUCT
TOTAL PRESSURE, ATA
HOURS ON STREAM
Tetralin fraction
Naphthalene fraction
Autofining of a mixture containing 33·3 %wt decalin and 66·7 %wt cycle oil
Autofining of a mixture containing 66·7 %wt decalin and 33·3 %wt cycle oil
(a)
(b)

FIG 6

TETRALIN FRACTION CONTENT AND NAPHTHALENE FRACTION CONTENT OF AUTOFINED DECALIN–CYCLE OIL MIXTURES IN RELATION TO TIME ON STREAM AND PRESSURE

dictated by the type and the quantity of naphthenes present in the feedstock. A feedstock of low naphthene content can be processed only at relatively severe operating conditions. Otherwise the dehydrogenation of the naphthenes will not satisfy the need of hydrogen for the desulphurization step. Consequent on the need for relatively high temperatures and low pressures for low naphthene containing feeds on stream, periods cannot be as long as in conventional hydrogen-consuming desulphurization processes. If it is desired to operate the autofining process at the higher pressures, which will prevent rapid decline of catalyst activity, the naphthene content of the feedstock should be high. If necessary, naphthenes of a naphthene-containing oil fraction can be added to the feedstock. High pressure autofining of an aromatic oil fraction with added naphthenes will be very unfavourable with respect to the hydrogen balance. In the first place, dehydrogenation of the naphthenes added to the oil fraction will be relatively low. Secondly, the aromatic rings of the compounds present in the oil fraction to be desulphurized will in part be hydrogenated into naphthenic rings.

ACKNOWLEDGMENT

The authors thank Dr E. C. Wessels for his help in these investigations.

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TWO NEW INSTRUMENTS FOR MEASURING THE AIR CONTENT OF OIL*

By A. T. J. HAYWARD†

SUMMARY

Two new instruments have been developed for measuring the air content of mineral oil. Both depend upon the admission of a sealed sample of oil to an evacuated de-aeration chamber and measurement of the rise in pressure resulting from de-aeration. Special sample bottles are described with which oil under pressure or under vacuum may be sampled, without bringing it to atmospheric pressure.

Both instruments are simple in construction, robust, and easy to operate. That which features a vibrating stirrer is very rapid in action and is considered to be more suitable for general use. The accuracy of an individual measurement has been found, in use, to be within ±3 per cent for both instruments.

INTRODUCTION

THE need for a simple instrument to measure the air content of oils has been apparent for some years. For instance, the air content of transformer oil is important because it affects electrical properties. The dissolved air content of oil supplied to pressure-fed lubricating systems and oil-hydraulic systems is important because it affects the liability of the oil to foam when low pressures are encountered. Measurement of the total air content (*i.e.* dissolved air + air bubbles) of the bubbly oil produced as a result of foaming may also be required.

There is no basic difficulty in measuring the air content of oil. Because the solubility of a gas in a liquid is proportional to the absolute pressure, air is insoluble in oil in an absolute vacuum. In theory, therefore, it is only necessary to apply a vacuum to a measured sample of oil, wait until all the dissolved air has come out of solution, and then measure the amount of air released. In practice, air is released from a bulk of oil very slowly, and it is necessary to accelerate the de-aeration process by agitating the oil or by spreading it out in a very thin film.

It has long been possible to measure accurately the air content of oil in a chemical laboratory, using fairly complicated and fragile apparatus. The engineer, however, needs a robust, portable instrument, simple enough to be used in workshops. An instrument was introduced in 1958 by Firth and Kane[1] to meet this need. This instrument represented a considerable advance over existing methods, and has since been used successfully in several engineering laboratories. It is, however, expensive to make and not as convenient to use as would be desirable. Efforts were therefore made to produce a simpler, cheaper, and more convenient apparatus. This note describes two new instruments which have been developed and for which patent applications are pending.

The new instruments, like some of their predecessors, employ the following basic method:

(*a*) A sample bottle, completely filled with oil, is sealed with a valve, and is then connected through that (closed) valve to a sealed chamber.

(*b*) The chamber is then evacuated, and the vacuum pressure measured.

(*c*) Next the valve is opened to admit the sample to the chamber where de-aeration occurs and produces a pressure rise.

(*d*) The pressure rise in the chamber and the ambient temperature are measured and from these figures the original air content of the oil is calculated.

For sampling purposes, oil may be considered as occurring in the following forms:

(i) Bubble-free oil at atmospheric pressure.

(ii) Bubble-free oil at high pressures inside a hydraulic circuit or machine.

(iii) Oil under vacuum in a hydraulic circuit or a laboratory apparatus.

(iv) Bubbly oil at or near atmospheric pressure.

SAMPLE BOTTLES

Special attention has been paid to the design of sample bottles, and two types have been developed.

The bottle for use with bubble-free oil at atmospheric pressure is shown in Fig 1. It is filled from a beaker and, to avoid entraining bubbles, the oil is poured down the interior wall. The cap is then screwed down (with the sealing valve open) and tightened to compress the O-ring which seals the cap to the bottle. Oil displaced by the central projection on the cap is forced up through the valve (Fig 1 (*b*)) when the cap is screwed down. Thus, when the sealing valve is closed, the bottle contains a sealed sample of oil, free from extraneous air.

The valve used is a high-quality diaphragm type, specially made for high-vacuum work, and has been found to be reliable. The neoprene diaphragms are

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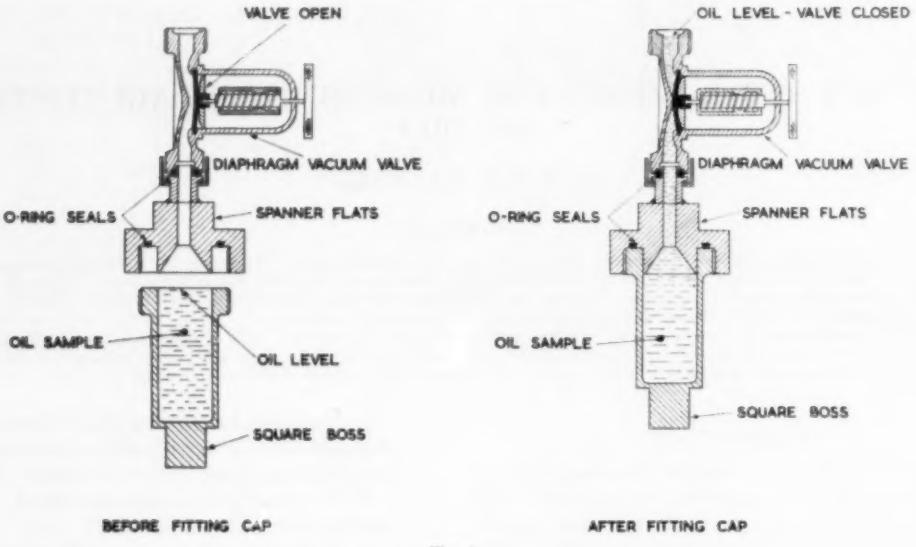

FIG 1

DETAIL OF OPEN-TYPE SAMPLE BOTTLE

unaffected by oil. Quick-release vacuum unions are incorporated which enable the sample bottle to be connected to the de-aeration chamber in a few seconds.

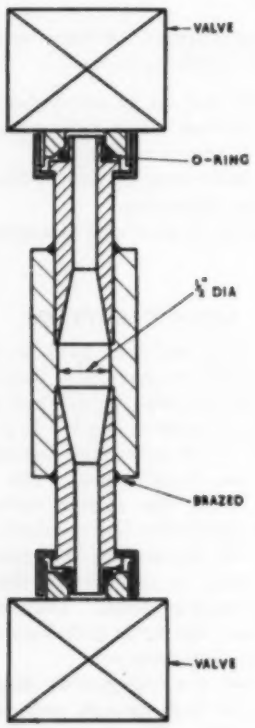

FIG 2

DETAIL OF PRESSURE-TYPE SAMPLE BOTTLE

To avoid premature release of dissolved air when oil under pressure is drawn off into an open vessel, the sample bottle shown in Figs 2 and 3 has been developed for use in conditions (ii), (iii), or (iv).

It consists essentially of a length of ½-inch bore pipe with a ¼-inch vacuum diaphragm valve at each end. These valves will withstand pressures up to 200 psi. If the pressure of the system is substantially greater

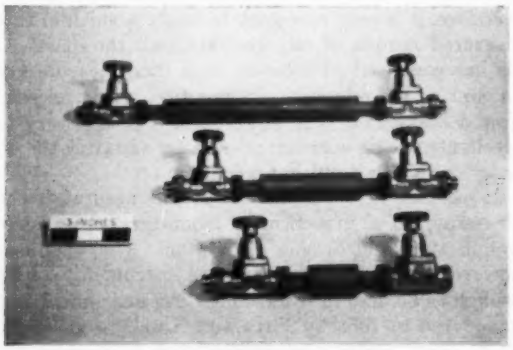

FIG 3

PRESSURE-TYPE SAMPLE BOTTLES

than this the pressure sample bottle must be connected to the system via a throttle valve and Bourdon gauge, so that the pressure in the bottle can be controlled at 150 to 200 psi. At this pressure there will be no tendency for air to come out of solution, unless the air content is at least ten times as great as the solubility of air in the same oil at atmospheric pressure; such high values of air content are extremely unlikely to occur in practice.

Three sizes of pressure-type sample bottles have been made, to cover the ranges 0 to 20 per cent, 20 to 40 per cent, and 40 to 80 per cent air content, the smallest bottle corresponding to the greatest air content.

When used for bubble-free oil at high pressures the bottle is held in a vertical position and connected at its lower end to the system being sampled. With the lower valve fully open and the upper valve slightly opened, it can be filled with oil at a pressure slightly below that of the system. By closing first the upper, then the lower valve, the sample of oil under pressure may be trapped inside.

When this bottle is used for oil in a system under vacuum it is connected to the system with a vacuum pump coupled to the outer end. In this way the bottle may be filled with oil at the pressure of the vacuum system.

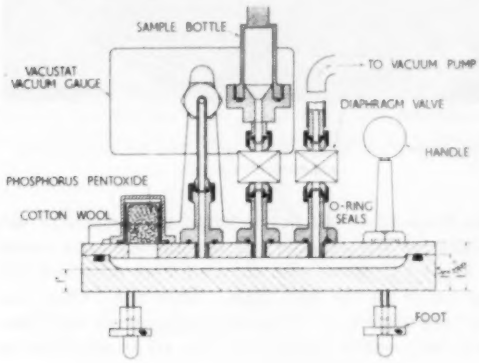

FIG 4

DETAILS OF CASSEROLE APPARATUS

For sampling of bubbly oil the use of an open-type sampling bottle gives unreliable results because of segregation; the pressure type bottle should be used with bubbly oil.

The bottle with both valves open is connected to the system and bubbly oil is allowed to flow through the bottle until equilibrium is established, when both valves are rapidly closed, trapping a sample of bubbly oil which is as nearly representative as it is possible to obtain.

THE CASSEROLE APPARATUS

This apparatus, shown in Figs 4 and 5, which derives its name from its shape, is a de-aeration chamber

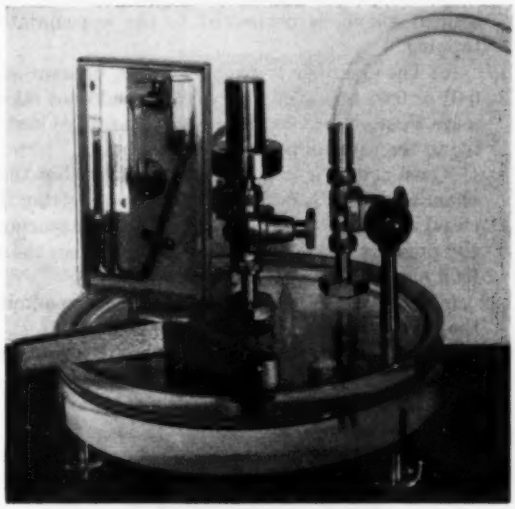

FIG 5

PERSPEX PROTOTYPE OF CASSEROLE APPARATUS

formed from a shallow dish, about 10 inches in internal diameter, with a flat spigoted lid resting on it and sealed by a neoprene O-ring. Three tappings penetrate the lid. One in the centre is used to connect the sample bottle; the second is connected through a vacuum valve to a rotary vacuum pump; and the third is connected by a short length of copper pipe to a "Vacustat" miniature McLeod gauge (range, 0 to 10 mm of mercury) which is fastened to the lid. No screws or fixings are used to hold the lid in place, since it is found that the weight of the lid, together with the force of atmospheric pressure, is sufficient to provide a vacuum-tight seal between the chamber and the lid.

It has already been shown[2] that, in an apparatus of this kind, an efficient dehydrating agent must be incorporated to eliminate errors caused by traces of water, which are often found in used oil. A pocket is therefore attached to the lid in which a pot of phosphorus pentoxide powder can be inserted to absorb any water vapour formed within the de-aeration chamber.

The base of the de-aeration chamber is fitted with three levelling feet. The base should be carefully levelled before use, so that when the oil is admitted to

the chamber it spreads evenly over the floor in a thin film. With the largest sample bottle the oil film thickness is about 0·022 inch.

The method of operation of the instrument is as follows:

(*a*) The moisture trap is filled with phosphorus pentoxide;

(*b*) The base is levelled and the lid placed in position;

(*c*) The sample bottle, previously filled as described above, is connected to the appropriate tapping.

(*d*) The chamber is evacuated to a pressure of 0·01 or 0·02 mm of mercury (this should not take more than about two minutes). The valve leading to the vacuum pump is then closed.

[Once a day it is advisable to check that the chamber is not leaking appreciably by letting it stand in this state for a few minutes and measuring the pressure rise. If the rate of rise is less than 0·05 mm/hr the leakage is negligible.]

(*e*) The sample bottle valve is opened to admit the oil to the chamber.

(*f*) When the pressure in the chamber reaches a constant value the air content of the oil is calculated from the formula:

$$\text{Air content} = 100 \cdot \frac{T_s}{P_s} \cdot \frac{V_c}{V_b} \cdot \frac{(P_2 - P_1)}{T} \text{ per cent by volume} \quad . \quad (1)$$

where

T_s and P_s are the standard temperature and pressure to which the air volume is to be reduced;

V_c is the volume of the de-aeration chamber;

V_b is the volume of the sample bottle;

P_1 is the initial pressure in the chamber, mm of mercury;

P_2 is the final pressure in the chamber, mm of mercury;

T is the ambient temperature in ° K (= 273 + ° C).

Substituting the values of $T_s = 273$ ° K and $P_s = 760$ mm of mercury at STP, equation (1) becomes:

$$\text{Air content} = 35{\cdot}92 \cdot \frac{V_c}{V_b} \cdot \frac{(P_2 - P_1)}{T} \text{ per cent by volume} \quad . \quad (2)$$

The casserole instruments now in use are made in mild steel, but a prototype was made in " Perspex," as shown in Fig 5. The use of this transparent material enabled the behaviour of the oil in the de-aeration chamber to be observed. On first being admitted the oil foamed violently, as shown in Fig 6, but the foaming died away after a minute or two. Curves showing the rate at which pressure rises after oil has been admitted are given in Fig 7 for three successive tests with one oil. These show that, with this oil, approximately 75 per cent of the dissolved air was released during this initial period of foaming. The remainder of the dissolved air was released by diffusion during the next 30 minutes.

The time taken for complete de-aeration, *i.e.* for the pressure in the chamber to reach a constant value,

FIG 6

APPEARANCE OF CASSEROLE APPARATUS AT INSTANT OF ADMISSION OF OIL

varies from oil to oil, but is found to be more or less constant for any one oil. It is therefore only necessary to plot pressure–rise curves for an oil when it is being tested for the first time. Once the " time constant " for a given oil is known, future tests with that oil are very easily performed; the oil is admitted to

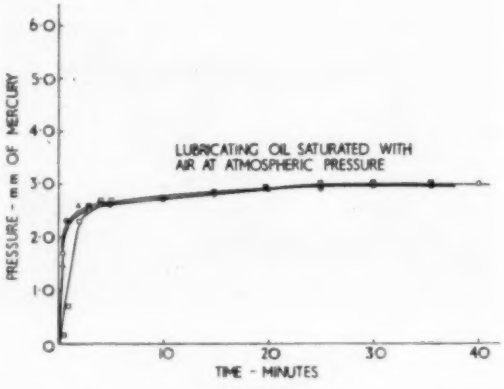

FIG 7

PRESSURE RISE IN CASSEROLE APPARATUS

the chamber and then the apparatus may be left unattended until the appropriate time has elapsed, when the final pressure reading is taken.

The time constants of six oils in this instrument have so far been found, and these are shown plotted against the viscosity of the oils in Fig 8. There

appears to be a tendency for air to be released more rapidly from the less viscous oils.

The great merit of this instrument is its extreme simplicity, which renders it inexpensive to construct and easy both to use and to clean out after use.

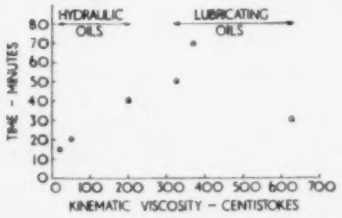

FIG 8

EFFECT OF VISCOSITY ON STABILIZING TIME—CASSEROLE APPARATUS

The one serious disadvantage is the length of time required for an air-content determination, which varies from less than 30 minutes in the case of a hydraulic oil of low viscosity to an hour or more for a viscous lubricating oil. However, the attention of the operator is required for only about 10 minutes of

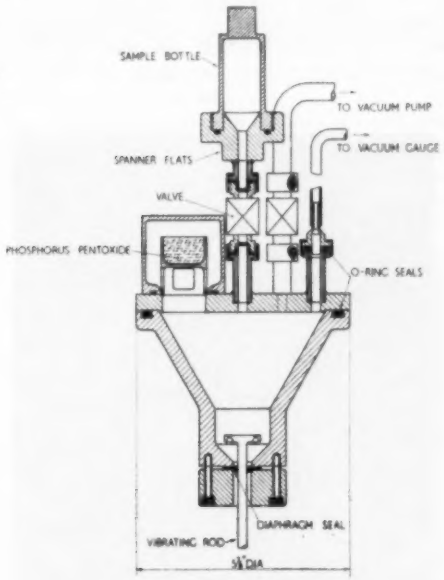

FIG 9

DETAIL OF FUNNEL APPARATUS

the test, and for the remainder of the time he can be available for other work. The number of measurements which may be carried out during one day with one instrument is, however, seriously limited. Great care must be taken to avoid leakage, since even a small leak can have a large cumulative effect during a prolonged test.

THE FUNNEL APPARATUS

This instrument, shown in Figs 9 and 10, has a de-aeration chamber in the form of a short, wide funnel. It is closed by a lid similar to, but smaller than, that of the casserole apparatus. This lid carries three tappings, as on the casserole lid, and a phosphorus pentoxide container of improved design is fitted. The bottom of the funnel is sealed by a flexible diaphragm of neoprene or silicone rubber penetrated by the shaft of a vibrating stirrer.

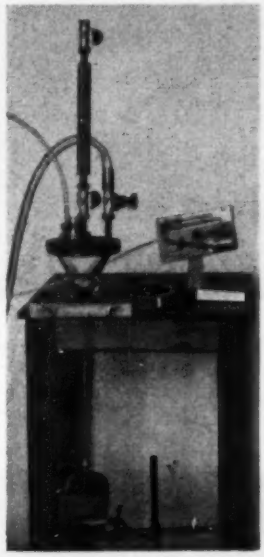

FIG 10

FUNNEL APPARATUS ASSEMBLED

The operating cycle is similar to that of the casserole apparatus, but the de-aeration process is greatly accelerated by the use of the vibrating stirrer. A series of photographs, taken at various stages during an actual test, is shown in Figs 11–16. The de-aeration process is complete in less than two minutes. A complete test, including the filling of the sample bottle and cleaning the apparatus after use, can be carried out in about 15 minutes.

The funnel apparatus is more complex and costly than the casserole instrument, due to the use of the vibrating stirrer (which costs about £30). This complication is, however, more than offset by the marked increase in speed of operation.

CALIBRATION OF BOTH INSTRUMENTS

In order to calibrate an instrument of this type the factor $\frac{V_c}{V_b}$ of equation (2) has to be determined. The volume of the sample bottle, V_b, may easily be

FIG 11
SAMPLE DISCHARGING INTO FUNNEL APPARATUS
2 sec after valve opening

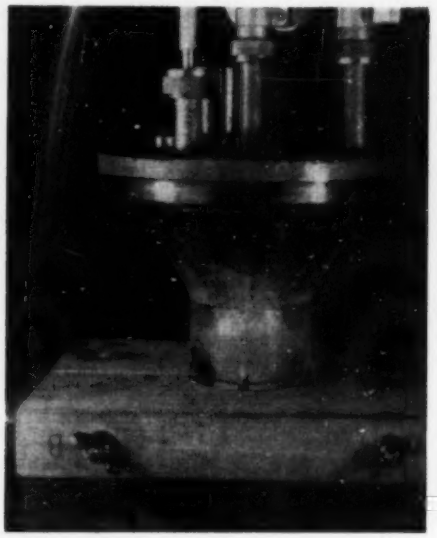

FIG 13
SAMPLE IN FUNNEL APPARATUS
21 sec after valve opening. Stirrer in motion

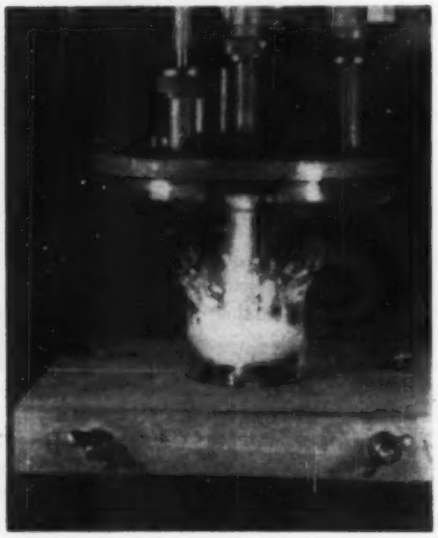

FIG 12
SAMPLE DISCHARGED INTO FUNNEL APPARATUS
13 sec after valve opening. No stirrer action

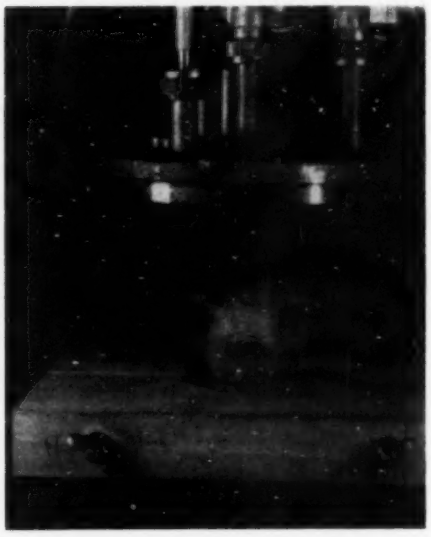

FIG 14
SAMPLE IN FUNNEL APPARATUS
33 sec after valve opening. Stirrer in motion

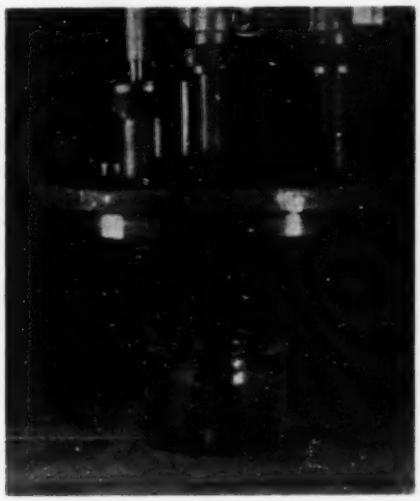

FIG 15

SAMPLE IN FUNNEL APPARATUS

54 sec after valve opening. Stirrer in motion, de-aeration almost complete

measured, but the volume of the chamber, V_c—which includes the volume of free space inside the phosphorus pentoxide vessel, the McLeod gauge, the connecting tubes, and the tappings—cannot be measured accurately by normal methods.

It is therefore preferable to use an experimental method of calibration, which was developed for use with the Firth–Kane apparatus but is equally applicable to the present apparatus. In this method,

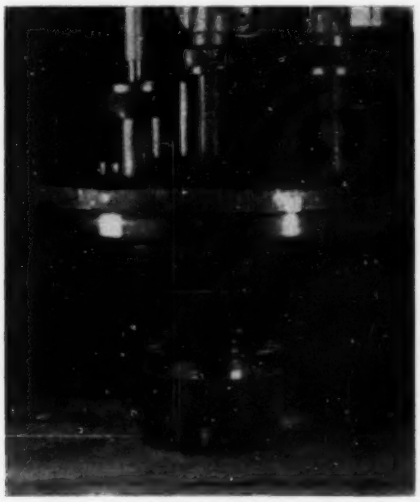

FIG 16

SAMPLE IN FUNNEL APPARATUS

130 sec after valve opening. Stirrer stopped, de-aeration complete

which has already been described in detail,[3] the sample bottle is first filled with dry air at a known pressure P_a (say, 250 mm mercury), and is connected to the de-aeration chamber with the connecting valve shut. The chamber is then evacuated to a pressure P_x (say, 0·02 mm mercury) and isolated from the vacuum pump. Finally, the sample bottle valve is opened and the resulting pressure in the chamber, P_y, is measured.

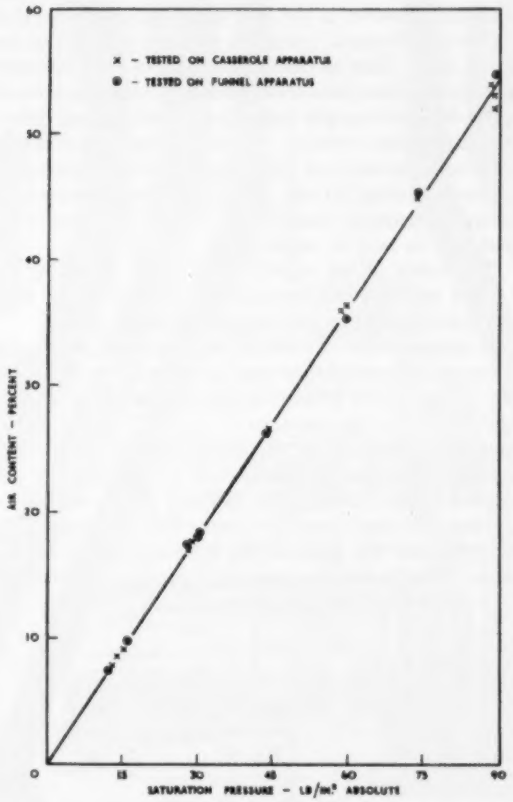

FIG 17

PRESSURE SOLUBILITY CURVE FOR AIR IN TELLUS 21 HYDRAULIC OIL

The required value is then given by the equation:

$$\frac{V_c}{V_b} = \frac{P_a - P_y}{P_y - P_x} \quad . \quad . \quad . \quad (3)$$

This calibration test can be carried out easily and quickly, and if the mean of several test results is taken as the value of V_c/V_b it should not be in error by more than about ± 1 per cent.

ACCURACY

According to Henry's law, the solubility of a gas in a liquid should be directly proportional to the absolute partial pressure of the gas. To find whether the

law holds for air and a typical hydraulic mineral oil, the oil was saturated with dissolved air at various regulated pressures up to 75 psi gauge. Several samples were taken from the oil at each pressure, and the air contents were measured using the casserole apparatus and the funnel apparatus in turn.

The air contents found are shown plotted against saturation pressure in Fig 17. The resulting straight line through the origin provides excellent confirmation of Henry's law. Of more interest in the present context, however, is the fact that with one exception all the experimental points lie within 3 per cent of the mean line. This is despite the use of two different de-aeration chambers (casserole and funnel) and about eight different sample bottles, all of which had different calibration factors. It can therefore be stated with some confidence that random errors (including calibration error) in any air-content measurement by either instrument should not exceed ± 3 per cent, and may in fact be much less.

The value of air solubility obtained from Fig 17, *i.e.* 8·9 per cent per atmosphere, agrees closely with the value quoted by the manufacturers of the oil.

A minor source of error in the measurements is the presence of residual dissolved air in the " de-aerated " oil. This will be present in the proportion of $P_2/760$ multiplied by the saturation value at atmospheric pressure, where P_2 is the final pressure in the de-aeration chamber in millimetres of mercury. P_2 is limited by the range of the McLeod gauge to 10 mm, so that the error from this source cannot exceed 1·25 per cent and will generally be in the region of 0·5 per cent. When extreme accuracy is sought, a correction can easily be applied which will eliminate error from this source.

CONCLUSIONS

Two new instruments have been developed which enable the air content of oil to be measured accurately. The instruments are simple to construct and easy to operate, and are sufficiently robust for everyday use in engineering laboratories and workshops. They are suitable for use with oil either at atmospheric pressure, under elevated pressures, or under vacuum.

The instrument known as the " funnel apparatus " is the more expensive, but it gives results very rapidly and is therefore regarded as the more suitable instrument for general use.

ACKNOWLEDGMENTS

The author is indebted to John Dallas and David Lawson for their assistance with the experimental work.

The work was carried out in the National Engineering Laboratory of the Department of Scientific and Industrial Research and is published by permission of the Director.

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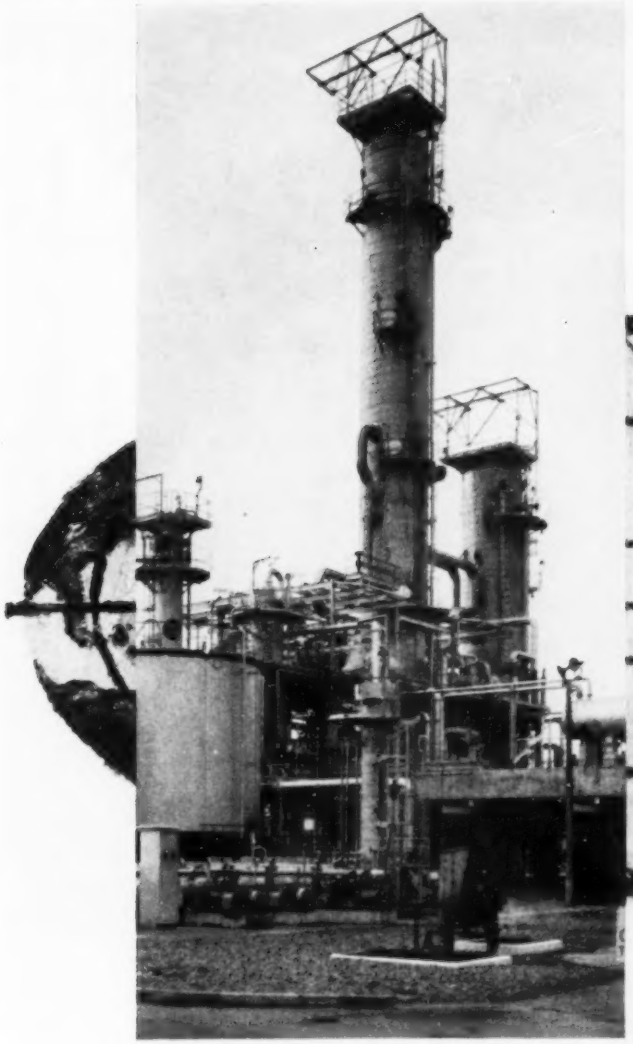

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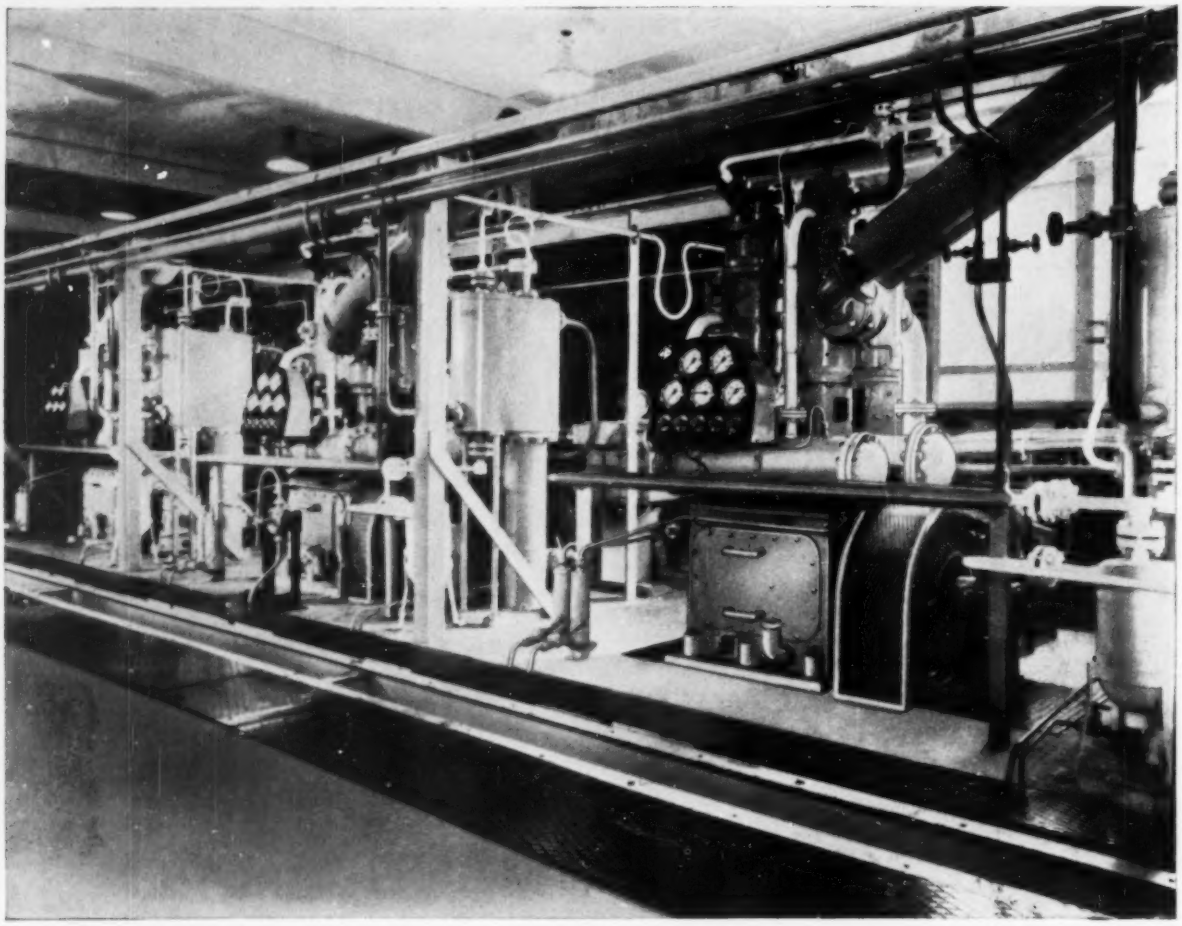

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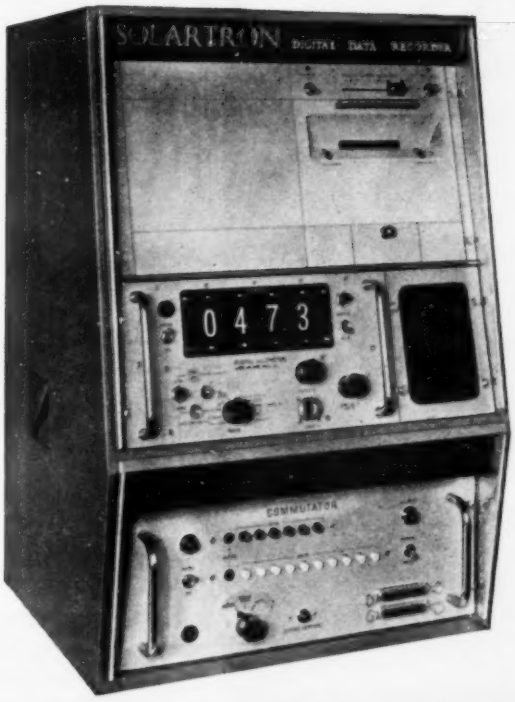

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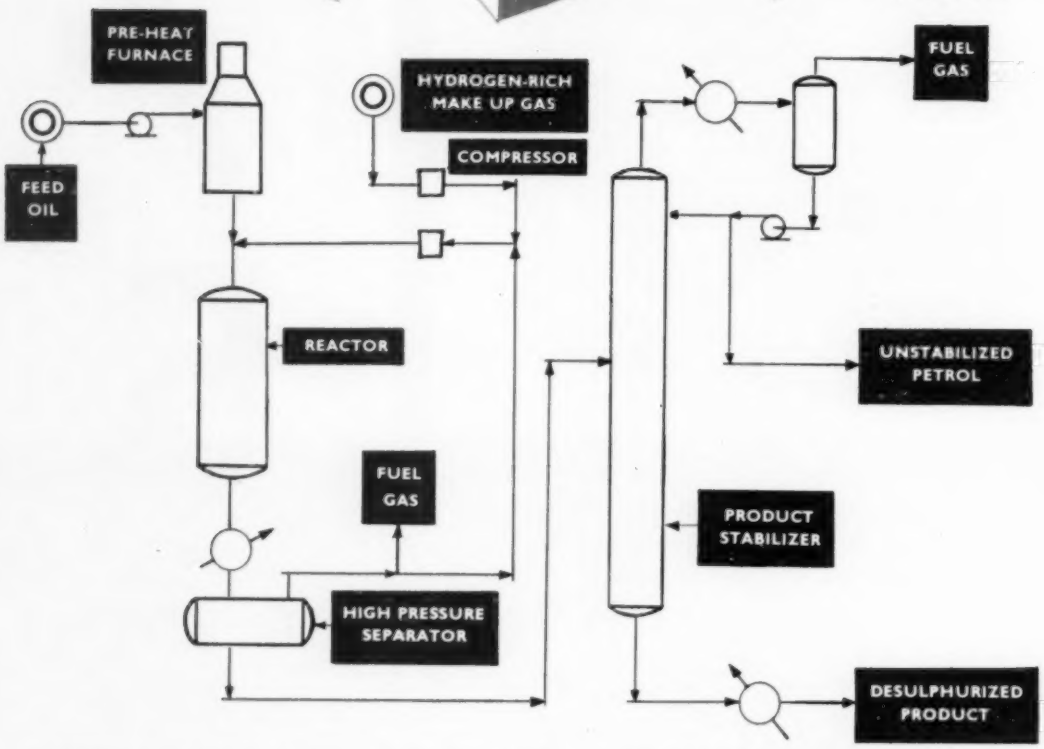

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	Feed	Prod.	Feed	Prod.
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ASTM Distillation, °F I.B.P.	340	370	420	430
E.P.	600	600	650	650
Sulfur, Weight Per centt	0.70	0.05	0.80	0.10
Weight Per cent Desulphurization	...	93.0	...	87.5
Fuel Oil, Vol. Percent on Feed	...	96.8	...	98.0
Unstabilized Petrol, Vol. Per cent on Feed	...	3.7	...	3.0

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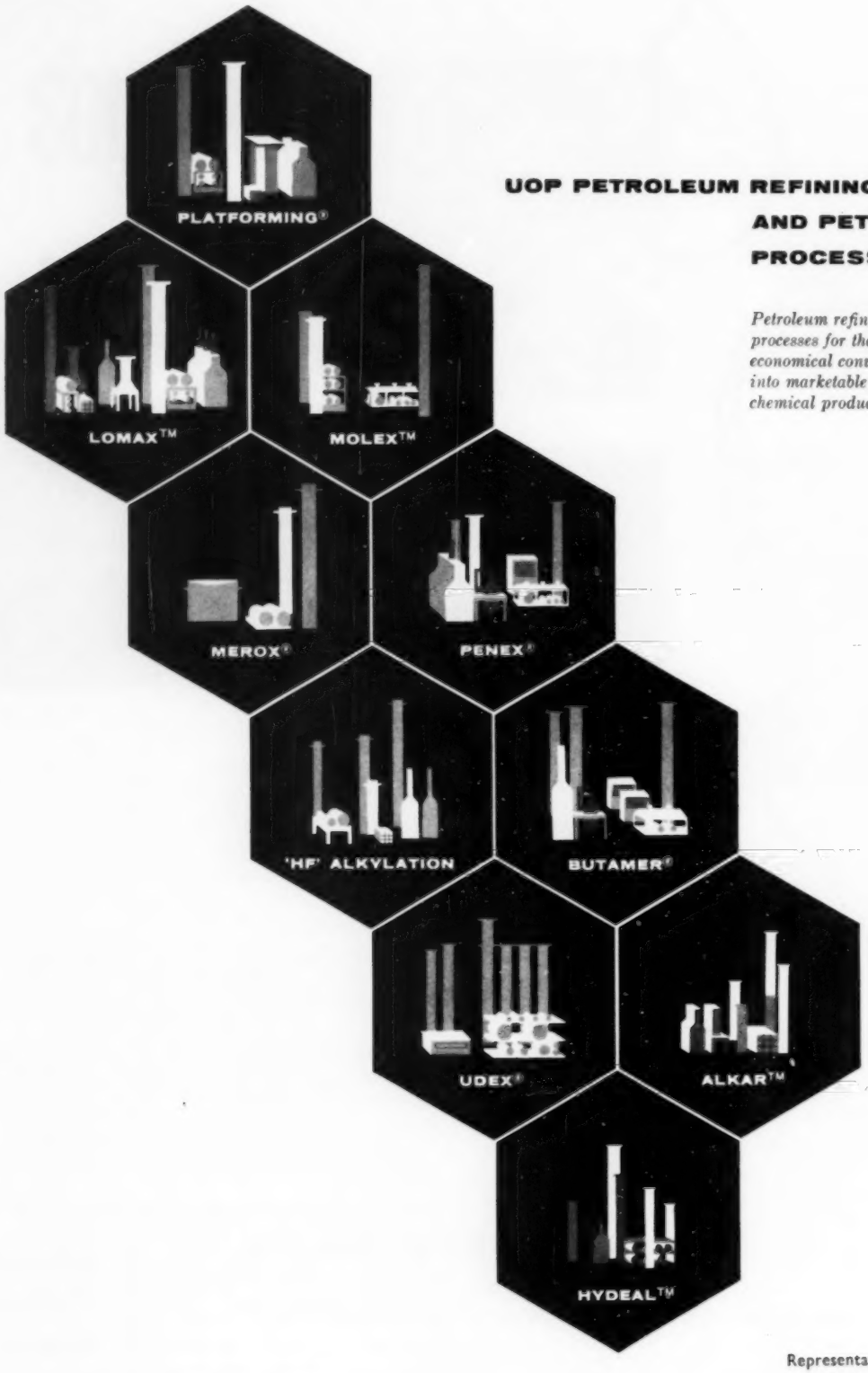

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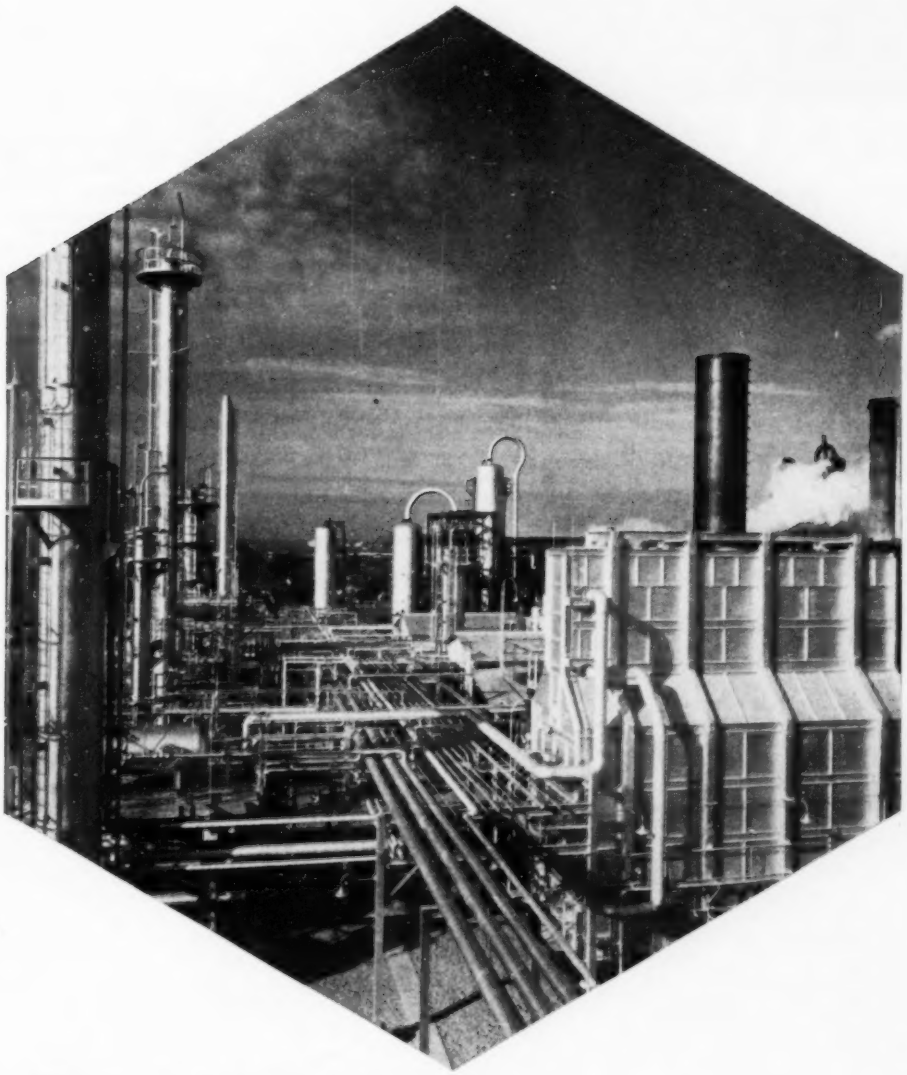

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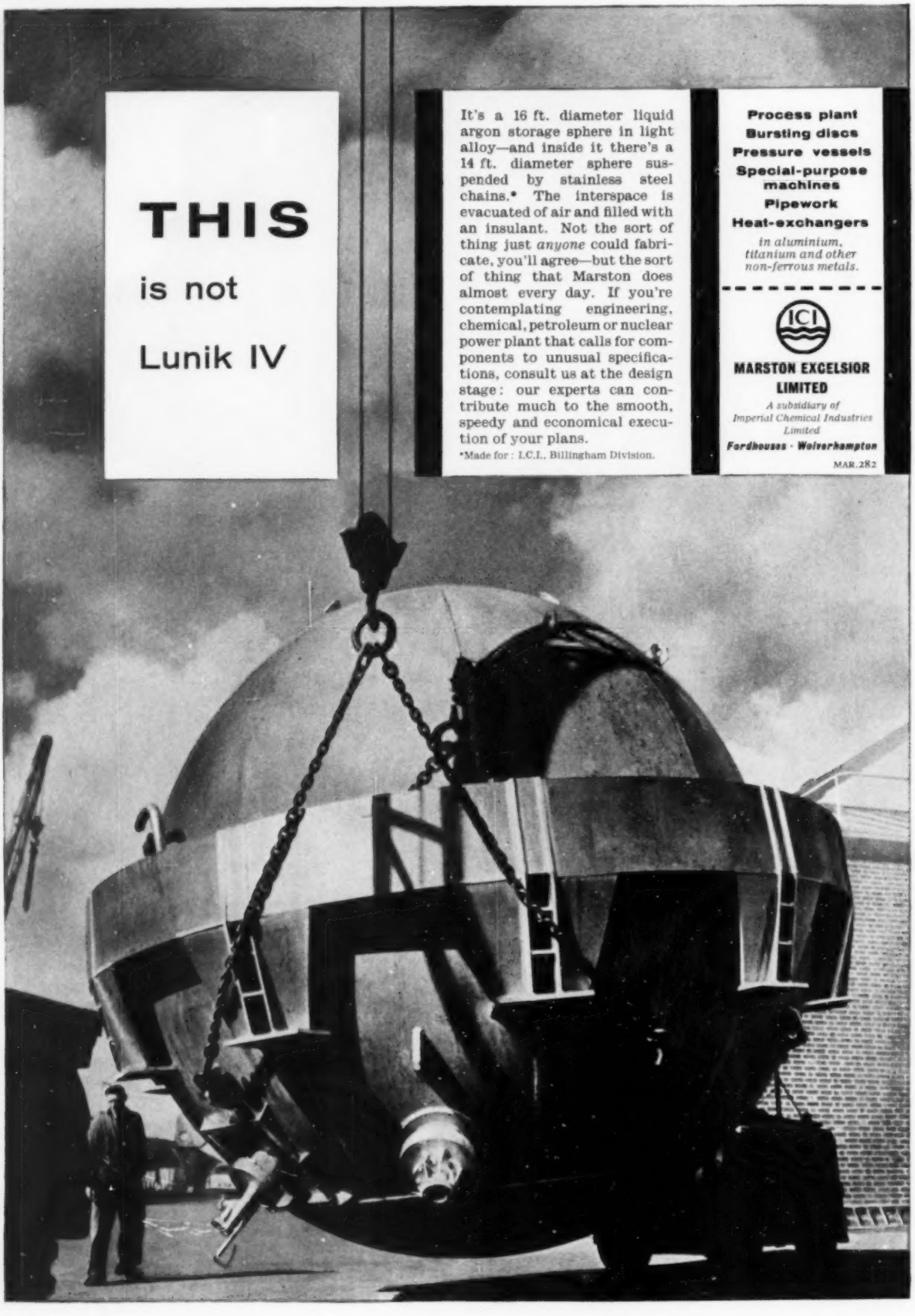
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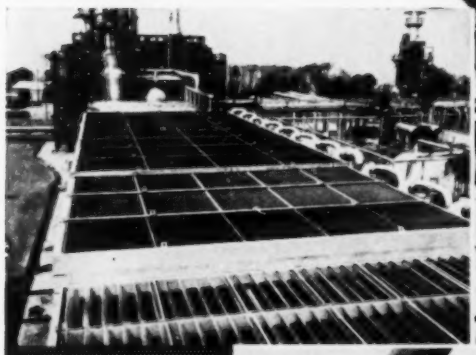

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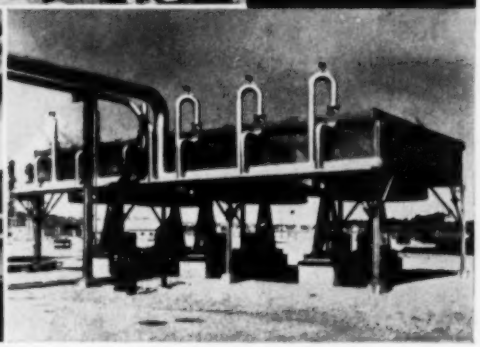

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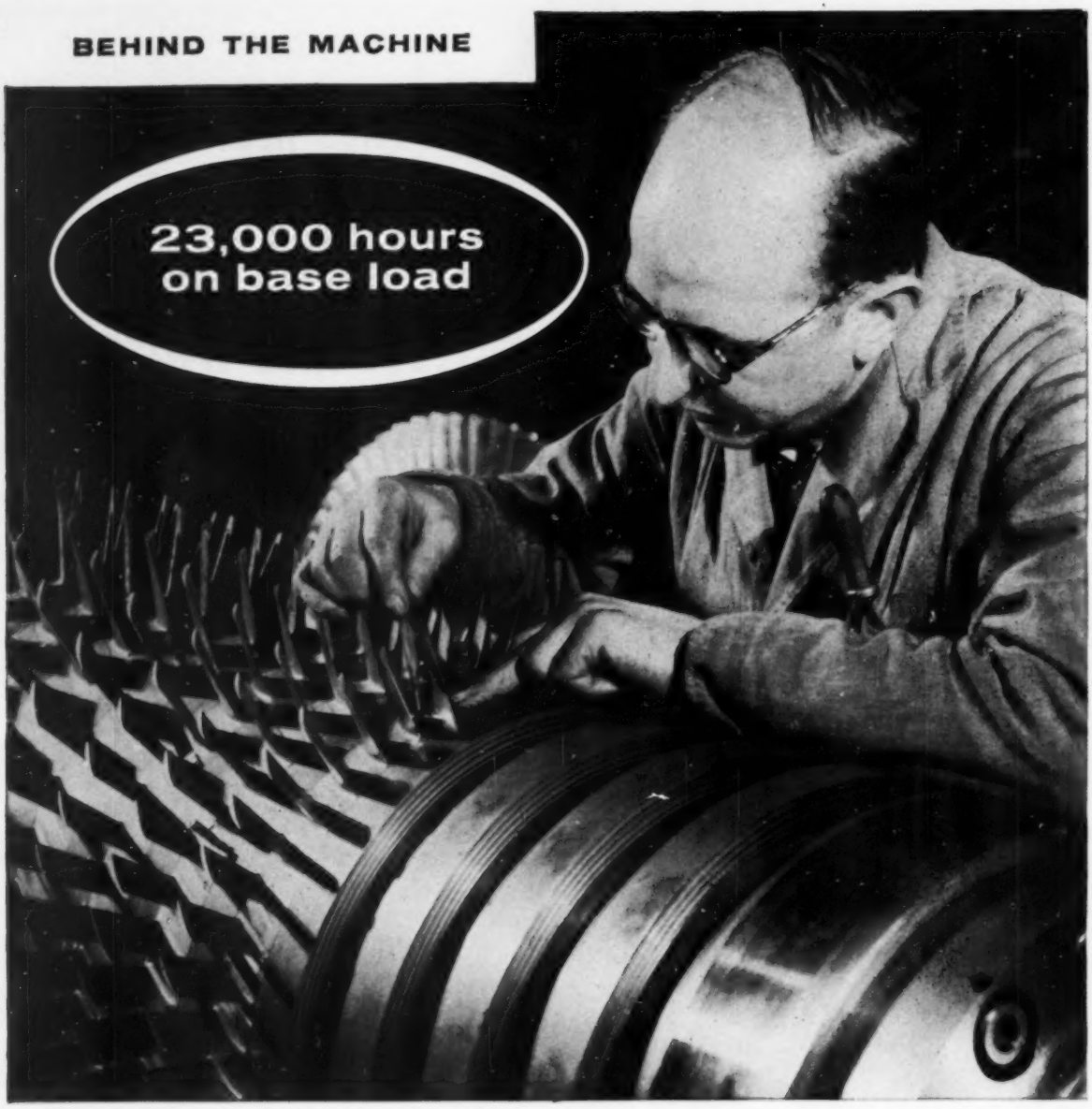

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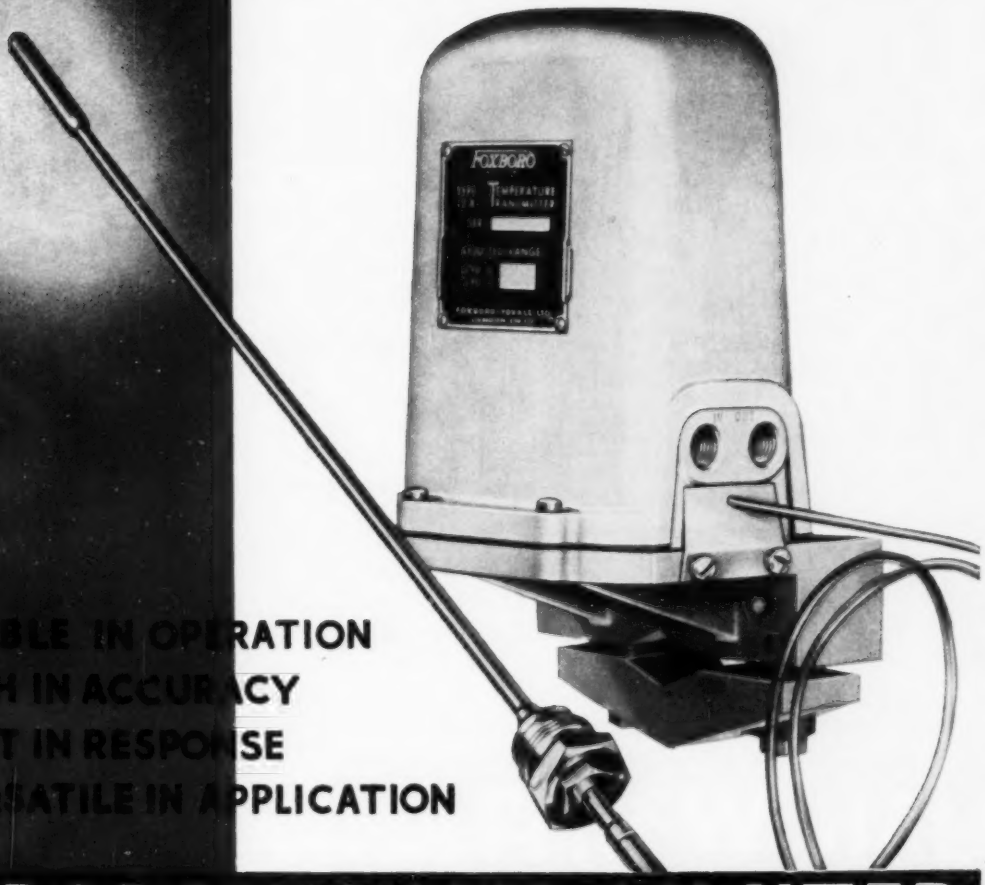

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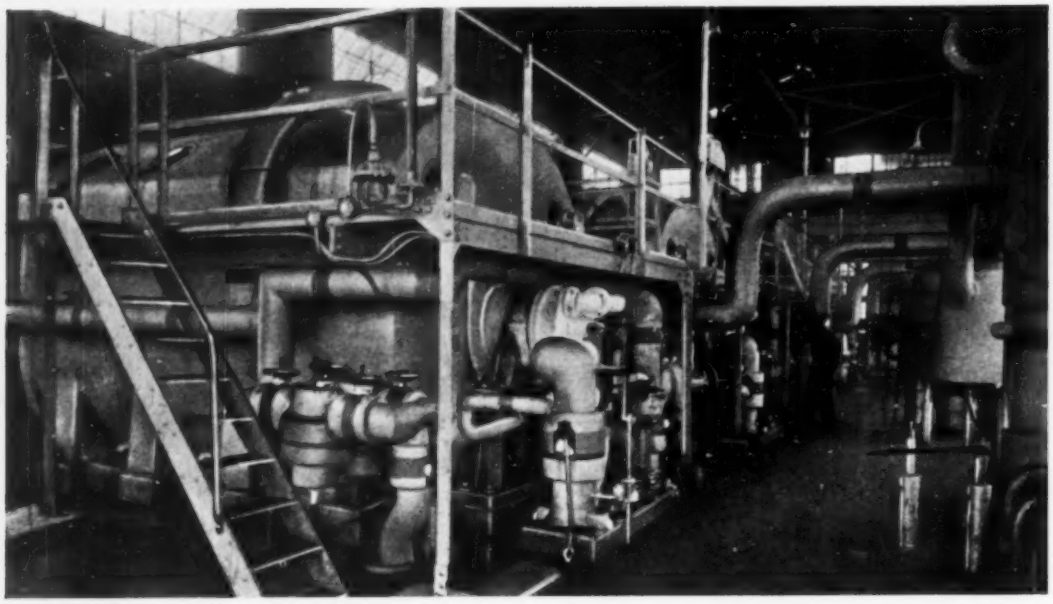

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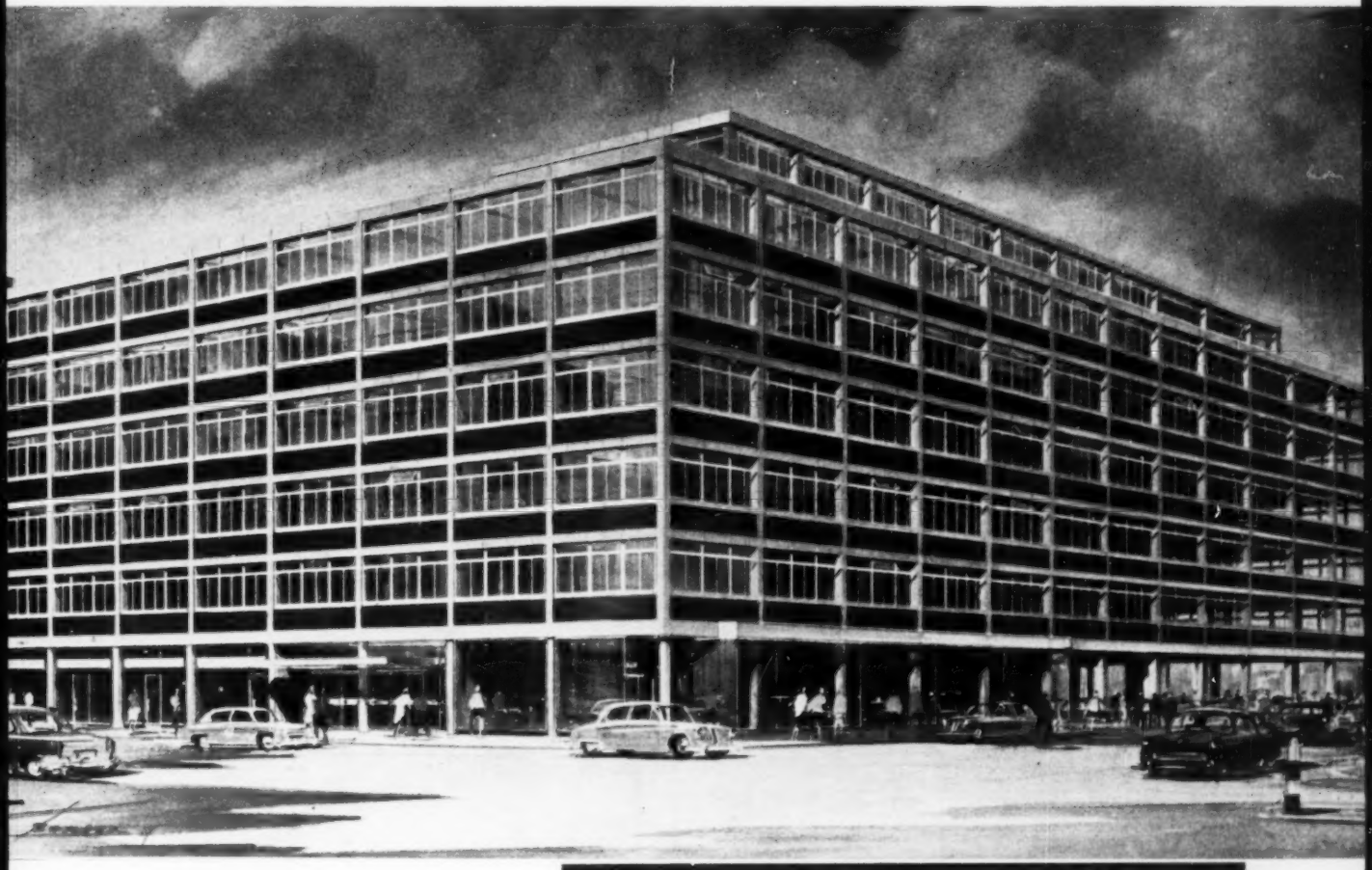

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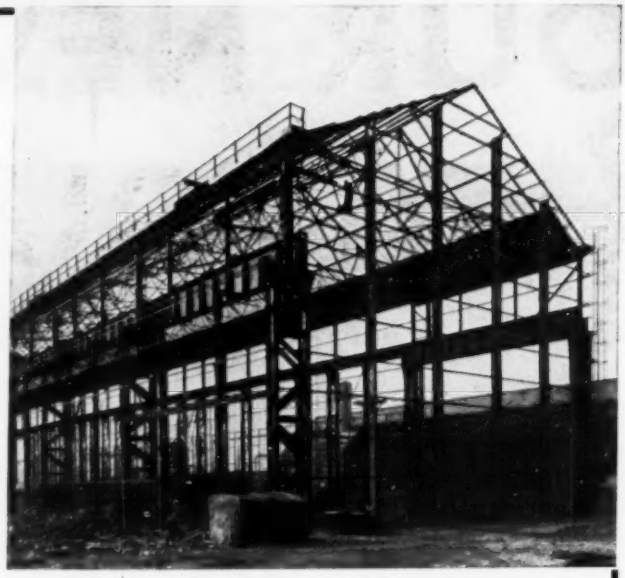

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